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VACUUM-INDUCTION MELTING, REFINING, AND CASTING OF URANIUM AND ITS ALLOYS

Ross J. Jackson

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SUBJECT DESCRIPTORS

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Casting Molds
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Fabrication
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Uranium Alloys

ROCKWELL INTERNATIONAL

AEROSPACE OPERATIONS
ROCKY FLATS PLANT
P. O. BOX 464
GOLDEN, COLORADO 80402-0464

MASTER

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VACUUM–INDUCTION MELTING, REFINING, AND
CASTING OF URANIUM AND ITS ALLOYS

Ross J. Jackson

ABSTRACT

The vacuum-induction melting (VIM), refining, and casting of uranium and its alloys are discussed. Emphasis is placed on historical development, VIM equipment, crucible and mold design, furnace atmospheres, melting parameters, impurity pickup, ingot quality, and economics. The VIM procedures used to produce high-purity, high-quality sound ingots at the U. S. Department of Energy Rocky Flats Plant are discussed in detail.

INTRODUCTION

Vacuum-induction melting (VIM) and casting of uranium is well-established, mature technology. Worldwide, this technology is used nearly exclusively to refine as-reduced metal and to convert this refined metal to useful shapes. It is also used extensively to reclaim scrap uranium metal by consolidating scrap to useful shapes. The resulting cast items may be ingots for subsequent rolling or extrusion operations or shapes that can be used more directly.

The vacuum-induction melting and casting method is also used nearly exclusively for preparing certain uranium alloys, such as the popular U-0.75 wt % Ti, U-2.0 wt % Mo, and U-0.2 wt % V alloys. Other popular alloys such as the U-6 wt % Nb or U-7.5 wt % Nb-2.5 wt % Zr alloys do not alloy well by vacuum-induction melting because of difficulty in dissolving the high melting point alloy additions. But, even in these cases, the vacuum-induction melting and casting method is used nearly exclusively to shape uranium feed for arc and electron beam alloying, as well as for scrap reclamation and consolidation.

A generalized flowsheet for vacuum-induction melting and casting of uranium and its alloys is shown in Figure 1. This report discusses various aspects of the development, operations, and logic of this flowsheet.

One intent of this report is to give an up-to-date, historical overview of vacuum-induction melting and casting of uranium and its alloys. The primary intent is to describe the best procedures for this process. The report is biased by the author's 23-year association with the uranium foundry at the Rocky Flats Plant near Golden, Colorado and a prior seven-year association with uranium casting at Iowa State College's Ames Laboratory. Hopefully, this report gives technical and historical insights that led to the procedures that today produce high-purity, defect-free uranium and uranium alloy castings.

URANIUM REDUCTION/REFINING/ CASTING DEVELOPMENT

1789-1939

Uranium was discovered by M. H. Klaproth[1] in 1789 in Berlin. His initial and subsequent papers[2-7] report the preparation of UO_2 which at the time was believed to be the metallic element rather than the oxide. The UO_2 was prepared by reduction of UO_3 in a carbon crucible. The UO_2, because of its metallic appearance and high density, was mistakenly considered by Klaproth to be the metal.

Uranium metal was discovered by E. Peligot[8] in 1856 in France. His initial and subsequent papers[9-12] report on a series of experiments in which UCl_4 was reduced to metallic uranium using the alkali metals, sodium and potassium. Aluminum was also used successfully. Density of the unconsolidated metal powder was determined to be 18.4 g/cm^3. One paper[11] by Peligot describes the reduction of UCl_4 with KCl and fresh-cut sodium in a porcelain crucible. He reports that the metal is smelted by rapidly heating to a high temperature

FIGURE 1. General Flowsheet for the Vacuum-Induction Melting and Casting of Uranium and Its Alloys

U CHARGE
1. Derbies
2. Top Crops
3. Solid Scrap
4. Briquettes

U CHARGE
Weigh, Alloy and Load into Crucible

CRUCIBLES
Brush Paint With Yttria

CRUCIBLES
Strip Lining

CRUCIBLES
Skull Burn-out

Oxidized Skull

Waste Disposal/ Recycle

Graphite Molds

VACUUM-INDUCTION FURNACE
1. 10-100μ
2. Coreless 960 Hz
3. Graphite Susceptor
4. Soak 1350 - 1450°C for 45 Min.
5. Stir (if necessary)
6. Bottom Pour, Power Off

MOLDS
Brush Paint with Yttria

INGOT BREAK-OUT

INGOTS
1. Cool
2. Weigh

Sawdust to Incinerator

MOLDS
Strip Lining

Molds

INGOTS
1. Saw Off Top (if necessary)
2. Scalp Sides (if necessary)

Recycle Top Crops

Briquette Chips and Recycle

NET SHAPE
Cups, Cones, Rods, Tubes, Plates, Etc.

INGOTS
1. Inspect Chemistry
2. Inspect Visually

FORGE
Cups, Cones

EXTRUDE
Rod, Tube

STRIP ROLL
Plate, Strip, Sheet

ROD ROLL
Rod

and that the unconsolidated metal can later be separated from the black slag by washing with water. Peligot stresses that oxygen must be excluded.

In the years between Peligot's metal reduction and 1938, numerous efforts were made to produce an unconsolidated metal of reasonable purity. A few efforts were also made to produce consolidated metal of reasonable purity. In general, the efforts were not very successful. In fact, in 1940, because of impure, unconsolidated metal, the melting point of uranium was thought to be 1850 °C. This is more than 700 °C higher than its actual melting point.

In the period 1856-1938, uranium metal reduction had been accomplished using aluminum,[8-16] calcium,[17-27] carbon,[28-30] potassium,[8-12,31,32] magnesium,[23,33-37] and sodium.[8-12,31,34,38-46]

In the same period, uranium metal reduction had been accomplished by the electrolysis of fused salts[37,47-55] and possibly by electrolysis in organic liquids[56] and acid solutions.[57,58] Uranium metal had also been prepared by thermal disassociation.[59-62]

Some efforts were made during the period 1856-1938 to purify uranium metal by distillation[16,24,25,57,63,64] and by sputtering thin films.[65-67] A few efforts were made to compact the unconsolidated metal by sintering[68,69] and by fusing unconsolidated metal in an electric arc.[44]

Most of these referenced papers reported success in their experiments. Generally, the metal was impure and unconsolidated. When metal was consolidated, it was generally impure and in small quantities. An impetus was needed for development of a new process or improvement of an existing one to produce a high-purity consolidated metal. The impetus was provided by Otto Hahn's discovery of uranium fission in December 1938 in Berlin.

The nuclear power implications of Hahn's discovery were soon recognized. By 1941, in the United States, nuclear physics experiments using uranium oxide showed that a self-sustaining nuclear reaction could be achieved by stacking natural uranium and graphite geometrically in a large pile. For the pile to work at the smallest possible size, the uranium and graphite would have to be as pure and as dense as possible. In late 1941, the task of finding a production process to produce large quantities of high-purity consolidated metal was assigned to Professor F. H. Spedding of the Iowa State College Chemistry Department.

1940-1945

The work leading to the present process began early in 1942 by scientists at Iowa State College in Ames, Iowa. The work was under the direction of F. H. Spedding, a chemistry professor. The present-day metal reduction and metal refining/casting process was developed there during 1942 and 1943. The process has stood the test of time. Considerable refinements have been made, but the process is basically the same today as it was in February 1943 in Ames.

In early 1942, it was well known that uranium metal could be prepared by one of several methods. Laboratory tests by the Iowa State team on some of the more promising methods for production were subsequently reproduced. However, these tests showed that the methods gave poor quality uranium metal or left much to be desired, process-wise, for a large-scale, low-cost operation. These conditions warranted an exploratory effort directed toward development of new methods for preparing uranium metal. A workable process employing the reduction of uranium tetrafluoride with calcium was the first major step in the desired direction. This process was soon followed by development of a workable method for producing uranium metal from uranium tetrafluoride with magnesium. In either case, the as-reduced metal was refined and cast by vacuum-induction melting.

When the program on atomic energy in the United States was expanded early in 1942, there were two American producers of uranium metal operating on a small scale. One producer (Metal Hydrides, Inc., Beverly, MA) used calcium hydride for reduction of uranium oxide. The mixture of UO_2 and CaH_2 was placed in a steel cylinder inside a retort, which was heated to 960 °C. After the reaction occurred,

the charge was allowed to cool under vacuum to eliminate the hydrogen. The product of the primary reaction was a sintered clinker, which was then crushed to a powder. This powder was subsequently leached with dilute acetic acid, washed, dried, briquetted, and sintered to form a fairly dense metal. This metal was rather impure and contained considerable oxide.

The other producer (Westinghouse Electric and Manufacturing Co., Pittsburgh, PA) employed an electrolytic process in which either KUF_5 or UF_4 was dissolved in a fused salt mixture of 80% $CaCl_2$ and 20% NaCl in a graphite crucible. This crucible served as the anode, and the uranium was electrolyzed out in the form of a granular deposit on a central molybdenum cathode. A temperature of about 900 °C was employed for the fused salt electrolyte to produce coarser, less pyrophoric metal. The uranium was subsequently leached, dried, briquetted, vacuum-induction melted, and cast to give good quality metal. The resulting small castings were made by vacuum self-induction heating of the uranium powder compacts supported in beryllia crucibles. Upon heating to high temperatures, the liquid uranium metal broke through its tough oxide skin and flowed through a grill into a beryllia mold.

Neither method met all the desired requirements as processes for large-scale, low-cost production of high-impurity uranium metal. Production was stepped up, however, by both methods beginning early in 1942. Plans for even larger expansion of the electrolytic plant were shaping up when the practicality of uranium tetrafluoride-calcium reduction followed by VIM refining and casting in graphite crucibles was demonstrated at Ames.

Several tons of uranium metal were produced in the chemistry laboratories at Iowa State College during the latter half of 1942 using the UF_4-Ca process. At the same time, a separate building on the campus was being remodeled and furnished to serve as a small production plant and a large-scale pilot plant. Meanwhile, efforts were also directed toward experimental development of the process employing magnesium in place of calcium.

All the uranium metal used in the successful Stagg Field nuclear-pile experiment (December 19, 1942)

came from three sources: Westinghouse in Pittsburgh, PA, Metal Hydrides in Beverly, MA, and the pilot plant at Ames. Most of the metal came from Ames and was produced by calcium reduction of uranium tetrafluoride. Before the end of 1942, Spedding and his associates concluded that none of these methods could produce uranium metal of the quality and quantity needed for full-scale plutonium-generating piles. Gambling on the ultimate superiority of magnesium reduction, the Ames group turned all its production efforts to that process in February 1943.

The key to the new process lay in the fact that magnesium reduction required a higher reaction temperature than the calcium method. The higher temperature was attained at Ames by placing the steel bomb containing the mixed charge of magnesium and uranium-tetrafluroide in a heat-soaking pit until the temperature of the charge was raised to the point of spontaneous ignition. The reaction developed sufficient additional heat to fuse the products, the dense liquid uranium metal collecting in the bottom of the bomb, and the magnesium-fluoride slag on top. The bomb was then allowed to cool and the solidified metal was removed as one large mass ("biscuit" or "derby"). The biscuits, weighing from 40 to 125 pounds in early 1943, were melted and refined in a vacuum-induction furnace and poured into graphite molds to form ingots about 4 inches in diameter and 13 inches long.

Experimental work at Iowa State College in 1942 had demonstrated that graphite crucibles could be employed for vacuum-induction melting of uranium. The melting point of uranium proved to be about 1130 °C (previously thought to be about 1850 °C); and at pouring temperatures to 1300 °C and even higher, the solubility of carbon in the uranium did not exceed a few hundred parts per million. In the early production pilot plant at Ames, uranium was induction melted in a graphite crucible with a grill in the bottom to catch oxide skins as the metal flowed into a heated graphite mold below.

The first uranium metal production plant at Ames employing the UF_4-Mg reduction and VIM refining and casting supplied metal for experiments and

charging atomic piles. It also served as a pilot plant where industrial technicians could inspect the process in operation and gain information for planning similar production plants elsewhere. The process was readily expandable, providing high-purity, defect-free uranium at a low cost. More than 1,000 tons of uranium metal billets were shipped from the Iowa State production plant before industry took over uranium metal production entirely.

So thoroughly did the Ames group investigate the magnesium process in early 1943 that no other method was considered for production plants constructed that spring. The largest was built at Niagara Falls, NY by the Electro-Metallurgical Company. Electromet produced the first ingot in its new plant in July 1943. A few days later, Mallinckrodt began production on the floor above its green salt plant in St. Louis. DuPont operated a small reduction facility at its Deepwater Point plant until August 1944, and Spedding expanded the pilot plant at Ames into a respectable production facility. Metal Hydrides and Westinghouse continued metal production during the summer of 1943. The Westinghouse electrolytic plant was shut down during the fall of 1943 after producing 65 tons of high-grade uranium metal. Metal Hydrides, DuPont, and Ames gradually shifted their operations to recovering ever-increasing amounts of uranium scrap and turnings from the slug fabrication plant at Hanford. This recovery operation was by VIM refining and casting. From time to time, there were minor fluctuations in the uranium metal supply lines, but this general pattern prevailed from the fall of 1943 until after the war.

1946 - Present

The Ames process for UF_4-Mg reduction and subsequent VIM melting, refining and casting of uranium is described in several post-war reports.[70-73] In the late 1940s and early 1950s, three large production uranium foundries were built in the United States. All were equipped with bottom-pour, vacuum-induction furnaces for the melting, refining, and casting of uranium. The three foundries were at the USAEC facilities at Oak Ridge, TN

(Y-12 Plant), Fernald, OH (Feed Metal Production Center), and Golden, CO (Rocky Flats Plant). The uranium processing plant constructed in the mid 1950s at Weldon Springs, MI did not contain a uranium foundry.[74] The Weldon Springs facility was designed to bomb-reduce a 1500-kg derby and extrude it directly to bar, rod, and tube. The Weldon Springs Plant was closed in 1963. The uranium foundries at Y-12, FMPC, and Rocky Flats are still in operation.

Since the metallurgy of depleted uranium was declassified by the U. S. Government in the mid-1950s, vacuum-induction melting, refining, and casting of uranium is well documented in the open literature. Perhaps the best single reference is a paper by Jaynes, Taub, and Doll.[75] This is an excellent 1957 state-of-the-art summary of the culmination of early VIM work at Los Alamos Scientific Laboratory and other laboratories and production facilities. It is devoted entirely to VIM refining and casting of uranium. Perhaps the best recent reference is a paper by Caddon, Jessen, and Lewis,[76] presented at the 1972 Vail Conference. It is an excellent 1972 state-of-the-art summary on uranium and uranium alloy melting at the Y-12 Plant. Subjects covered include induction, arc, skull, electroslag, inductoslag, and plasma-arc melting. Many other excellent papers on VIM refining and casting of uranium and its alloys are in the open literature.

Several monographs[77-81] on uranium metallurgy contain discussions on vacuum-induction melting and casting. Probably the most informative discussion is by Harrington and Ruehle[77] who discuss VIM casting of unalloyed uranium at the USAEC Feed Metal Production Center at Fernald, OH and VIM casting of alloys. Coreduction and arc-melting of alloys are also discussed. The preparation of alloys of U-Al, U-C, U-Mo, U-Nb, U-Si, U-Ti, U-Zr, and U-Nb-Zr by VIM casting are described. Wilkinson[78] gives a generalized treatment of VIM casting of uranium and its alloys. Wilkinson[79] later discusses the VIM casting of U-Mo and U-Nb alloys. Gittus[80] gives a discussion of VIM casting in England (Springfield Uranium Processing Facility), France, and the United States. Galkin et al.[81] gives an informative discussion of VIM melting and casting of uranium in the USSR.

The open literature contains papers related specifically to mold materials,[82] mold coatings,[83-88] mold design,[21,89] mold heating,[90,91] solidification mechanisms,[92-94] segregation,[95] purification,[96-99] factors affecting ingot quality,[21,100-103] centrifugal casting,[104] injection casting,[105] large castings,[106] casting natural uranium,[107] casting enriched uranium,[108] theoretical considerations in casting[109] and in computer applications.[89,93,94]

Papers describing vacuum-induction melting and casting procedures at uranium foundries in the United States have been published by Battelle Memorial Institute[110] at Columbus, OH, the Feed Metal Production Center[111,112] at Fernald, OH, Hanford Works[113] at Richland, WA, the Los Alamos Scientific Laboratory[75] at Los Alamos, NM, and the Y-12 Plant[76,114] at Oak Ridge, TN.

Papers discussing or mentioning work on vacuum-induction melting, refining, and casting of uranium were published by France,[115] Germany,[116] Italy,[117] Russia,[81,118,119] and the United Kingdom.[80,84,120-122] A paper on electric-resistance-heated vacuum melting of uranium was published by Belgium.[123]

Articles have also been published relating to the vacuum-induction melting and casting of alloys of U-Al,[77,124,125] U-C,[77] U-Cr,[126] U-Mo,[77,79,125,127] U-Nb,[77,79,102,128-131] U-Si,[77,125] U-Ti,[77,95,101,125,130] U-V,[125] and U-Zr.[77,104,129,132]

VACUUM-INDUCTION MELTING

Induction Heating Development

In 1831, Michael Faraday discovered the phenomenon that allows induction heating. His experiment, in which two coils of wire were wound on an iron ring, provided the first transformer on record and the first realization of the phenomenon of electromagnetic induction. The successful operation of such a mechanism or device to bring about transformation of energy by induction required alternating currents; and in 1840, automatic mercury or platinum contact interrupters (called spark gaps) had come into use. Shortly after 1850, a large number of patents were issued in the United States and abroad, covering

high-frequency apparatus for heating of metals. General interest and use at that time was for melting, wherein a metallic crucible was heated by induction and the heat of the crucible was transmitted to the charge by conduction and radiation (coreless heating).

The first full-scale attempt to induction melt metal occurred about 1890 in a ring-type furnace developed in Sweden by Kjellen. Production records are scant, and the actual performance and use of this furnace is not known.

Progress was slow, and it was not until 1917 that recorded successful melting on a production basis was demonstrated. This occurred at the Bridgeport Brass Company in the United States. Electrical energy was induced in a submerged loop of molten metal by linking it through coils built into the furnace, taking power from 60-Hz energy (core-type heating). The first production-scale *vacuum*-induction melting furnace was built by Heraeus of Hanau, West Germany (near Frankfurt). This furnace was complemented by two additional furnaces in 1926, each with a four-ton charge capacity.

The main growth in vacuum-induction melting and casting began shortly after World War II. This was due to development of efficient vacuum pumps and the high-quality material requirements necessary for rapidly developing nuclear and aerospace industries.

In the early 1900s, E. F. Northrup experimented with coreless induction melting. He perfected and patented many of the furnaces in use today and developed the first production coreless furnace installation in the United States. Early Northrup furnaces were spark-gap converter powered; later, with help from the General Electric Company, high-frequency motor generators to 960 Hz became available. Still later, the frequency range of rotating converters was extended in stages to incorporate 2000, 3000, 10,000, and 20,000 Hz.

Bearing problems and associated high repair costs led to the search for static devices to power induction furnaces. Spark-gap converters were very limited in maximum power output and, in

general, had maintenance and electrical problems limiting their use. In the early 1940s, a mercury hydrogen converter was developed by Allis Chalmers Company and sold in ratings to several hundred kilowatts at frequencies ranging from 800 to 1500 Hz. A converter based upon a saturable transformer was developed by Ajax Electrotherm Corporation in the 1950s. This system tuned to and used the third harmonic to work into a furnace coil at 180 Hz. Later, Inductotherm piggybacked a saturable tripler on a saturable tripler and produced 540 cycles for smaller furnaces and applications requiring higher frequency.

The 1970s and 1980s were truly the decades of solid state for the induction industry. Silicon-controlled rectifiers (SCR) in solid-state systems became all-pervasive, to the point where new induction heating and melting installations at any frequency will be SCR controlled. Solid state has made possible any frequency from 60 Hz to 50 kHz on single power blocks up to four megawatts. The takeover is primarily based on economics with initial costs in some cases less than half that of rotating converters. Flexibility in frequency and control, plus high-conversion efficiency, also make solid-state SCR conversion the most attractive system. The future of induction melting and heating is closely related to the future of solid-state circuitry.

The historical development of the vacuum-induction melting of uranium began in early 1942. The vacuum-induction melting, refining, and casting method was used by the Iowa State team to consolidate, purify, and shape their calcium-reduced metal in 1942. The merits of vacuum-induction melting were soon recognized such that the early uranium production foundries at Iowa State College and Electromet (Niagara Falls, NY) used bottom-pour, vacuum-induction melting and casting techniques. The three large USAEC uranium production foundries that came on line in the late 1940s and early 1950s were equipped with bottom-pour VIM equipment. Other privately-owned uranium production casting facilities have come on line since that time, i.e., Nuclear Metals Inc., of West Concord, MA and Barnhill County, SC (actually a subsidiary called Carolina Metals), National Lead Company in Albany, NY (presently defunct and under USDOE guardianship), and Tennessee Nuclear Systems of Jonesboro, TN (a subsidiary of Aerojet Ordnance). All were designed with bottom-pour VIM furnaces.

Induction Heating Principles

Induction heating is well-documented technology. Numerous monographs[133-138] have been published on the subject. A short treatment of the principles involved in induction heating is that of Leatherman and Stoats.[139]

Induction heating is an electrical method of heating metals or conductive materials for processes such as melting, surface hardening, brazing, soldering, welding, tempering and annealing, forming and extrusion, shrink fitting, and many others.

The induction heating circuit is, essentially, a transformer. The inductor, or work coil carrying the alternating current is the transformer's primary and the material to be heated becomes the secondary when placed within the loop formed by the inductor. There is no physical contact or connection between the work and the work coil. The magnetic field created in the work coil induces a flow of current in the material to be heated. Heat is generated by the induced flow of current and the material's electric resistance to the current, i.e., the I^2R losses.

In addition to I^2R heating (frequently referred to as induced-current or eddy-current losses), another form of heating takes place in ferromagnetic materials known as hysteresis losses. This is a result of "molecular friction" caused by magnetizing, demagnetizing, and remagnetizing the material in opposite directions by the alternating current. It explains why ferromagnetic materials heat faster below the curie temperature with induction heating than non-magnetic materials. In non-magnetic materials, only the eddy-current losses exist.

Commonly used frequencies for induction heating are 60-Hz (commercial power lines), 180, 540, 1000, 3000, and 10,000 Hz (motor generators or static power supplies), 20,000 to 600,000 Hz

(spark gap or vacuum tube oscillators), and 1,000,000 to 500,000,000 Hz (vacuum tube oscillators). A hertz is a unit frequency equal to one cycle per second. There is a cost advantage in using the lowest frequency that will do the job. For example, high-frequency operations are generally three to 15 times more expensive than 60-cycle operations. Sixty-cycle (line frequency) heating requires no frequency conversion equipment and accounts for its large economic advantage. Generally, motor generators and static power supplies (180 to 10,000 Hz) are less expensive (capital plus operational costs) than spark gap or vacuum tube oscillators (20,000 to 600,000 Hz), which in turn are less expensive than vacuum tube oscillators operating in the 1,000,000- to 500,000,000-Hz range. Solid-state SCR systems are about half the cost of non-solid-state frequency generating systems.

Different frequencies are necessary because of a phenomenon known as the "skin effect." This occurs when current induced in the material being heated is concentrated into a thin section on the periphery of the material. Heat travels from the outer skin by conduction to the inside of the piece. The depth of this initial current penetration varies inversely as the square root of the frequency of the current. The lower the frequency, the greater the depth of penetration. Therefore, thorough heating, as used for forging or melting, requires lower frequencies than those used for surface heating applications such as case hardening. The skin effect is an important consideration in selecting proper equipment for any given application.

The size of the piece to be heated must be considered along with the skin effect, i.e., the depth of direct heating penetration. When depth of penetration exceeds half the thickness of the piece to be heated, there is cancellation or bucking of the currents at the center of the piece with resultant loss of efficiency. This is the effect that limits the minimum size of the piece that can be efficiently heated at a given frequency.

The depth of current penetration as a function of frequency for uranium and several other metals is estimated in Table 1. The depth of penetration was calculated using the well-known relation:

$$\text{Penetration Depth (inches)} = 3.15 \sqrt{\frac{\rho}{\mu \nu}}$$

Where ρ is the resistivity in micro-ohm inches of the material at the temperature to be achieved, μ is the permeability of the material, and ν is the frequency of the power supply. For the calculations in Table 1, the permeability of the material was assumed to be one.

VIM EQUIPMENT AT ROCKY FLATS

Overview

The depleted uranium production foundry at the USDOE Rocky Flats Plant became operational in

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TABLE 1. Depth of Current Penetration as a Function of Frequency for Uranium and Other Metals. The values are averages for the working temperature range.

Metal	Resistivity Micro-ohm (inches)	Depth of Penetration (inches)				
		60 Hz	540 Hz	1000 Hz	3000 Hz	10000 Hz
Aluminum	2.24	0.61	0.20	0.15	0.086	0.047
Copper	1.8	0.55	0.18	0.14	0.078	0.043
Mild Steel	(below Curie Temp.)	0.50		0.12	0.060	0.030
Mild Steel	(above Curie Temp.)	2.60		0.65	0.039	0.20
304 SS	45.0	2.74	0.91	0.67	0.39	0.21
Titanium	49.0	2.86	0.95	0.70	0.40	0.22
Uranium	17.5	1.71	0.57	0.42	0.24	0.13

1953. The original foundry contained 12 Stokes vacuum-induction melting furnaces. All the power supplies were motor generators. The vacuum systems were mechanical pumps backed by oil diffusion pumps. The furnaces were top-loading, bottom-pour in design. Eight of the 12 furnaces had 66-inch inside diameters. The other four furnaces had 36-inch inside diameters. The larger furnaces were rated at 250 kW and 960 Hz. The four smaller furnaces were rated at 100 kW and 3000 Hz.

Two of the original 12 furnaces (36-inch i.d. units) were removed in the early 1960s to make room for a beryllium machine shop. Two 36-inch i.d. units were removed and scrapped in 1982 and 1983 to make room for a larger electroplating shop.

Major renovations of the eight remaining large furnaces occurred from 1985 to 1987. The vacuum pumping efficiencies were doubled by new mechanical roughing pumps and by replacing the oil diffusion pumps with Roots blowers. The motor-generator power supplies were replaced with solid-state SCR converters. Several new induction coils were obtained. New temperature and pressure monitors, controllers, and recorders were installed.

The remaining eight production VIM furnaces are in their original locations. They require three floors for operation. However, all metallurgical operations are conducted on the top (third) floor. Only maintenance operations are carried out on the first two floors. Furnace controls, power supplies, and vacuum systems are on the top floor. Loading and unloading operations occur on the top floor. The middle floor contains the solid-state converters, bus bar, mechanical roughing pumps, Roots blowers, and most of the furnace shells. The lower floor contains the furnace load lifting rams, hydraulic pumps, circulating water pumps, and water makeup tanks.

Furnace Shell and Coils

A cutaway view of a Rocky Flats VIM furnace is shown in Figure 2. All furnaces are top-loading and bottom-pour in design with identical 66-inch-diameter shells, pumping systems, power supplies, controls, and monitoring systems. Five furnaces have 42-inch-diameter coils; three have 28-inch-diameter coils. All the coils are four feet in height. The three furnaces with 28-inch diameter coils were replaced with 42-inch-diameter coils in 1987-88 to increase foundry melt capacity. The 42-inch coil accepts a 35-inch-diameter crucible and mold, while the 28-inch-diameter coil accepts a 23-inch-diameter crucible and mold. Figure 2 shows the arrangement with the smaller diameter coil.

The furnace shell is of double-wall construction. The outer wall is carbon steel. The inner wall is a non-magnetic grade of stainless steel. A non-magnetic material for the inner wall prevents inductive coupling and subsequent heating of the shell. The space between the inner and outer walls serves as a water jacket for cooling purposes. The furnace top opening is for loading and unloading the crucible and mold. The bottom opening is for maintenance, repair, and cleaning major metal spills. The openings are sealed with O-rings. The top has a sight glass to view crucible conditions and to read the optical temperature of the metal. There is an opening in the middle shell for the stopper rod lifting mechanism. There are also thermocouple lead wires, openings, a sight glass shutter opening, a power port opening to provide bus bar inlet for the induction coil, a high-vacuum port opening to facilitate evacuation of the furnace, and a ventilation port opening to downdraft the furnace during hot-stripping operations. Two 42-inch-diameter coil furnaces have an opening in the lid for a mechanical plunger stirrer. All openings are sealed with O-rings.

The induction coil is fabricated from copper tubing providing an electrical function and a mechanical function. The electrical function is to act as an inductance; the mechanical function is to cool the induction coil. One-inch-diameter tubing is flattened to an elliptical shape and wound in the form of a helix to the proper diameter with the proper number of electrical turns to form the coil. Each 42-inch-diameter coil consists of six copper tubes each having four turns within four feet.

Between the coil and the graphite crucible mold is a firebrick lining. There is about one-inch clearance between the firebrick and coil and between the
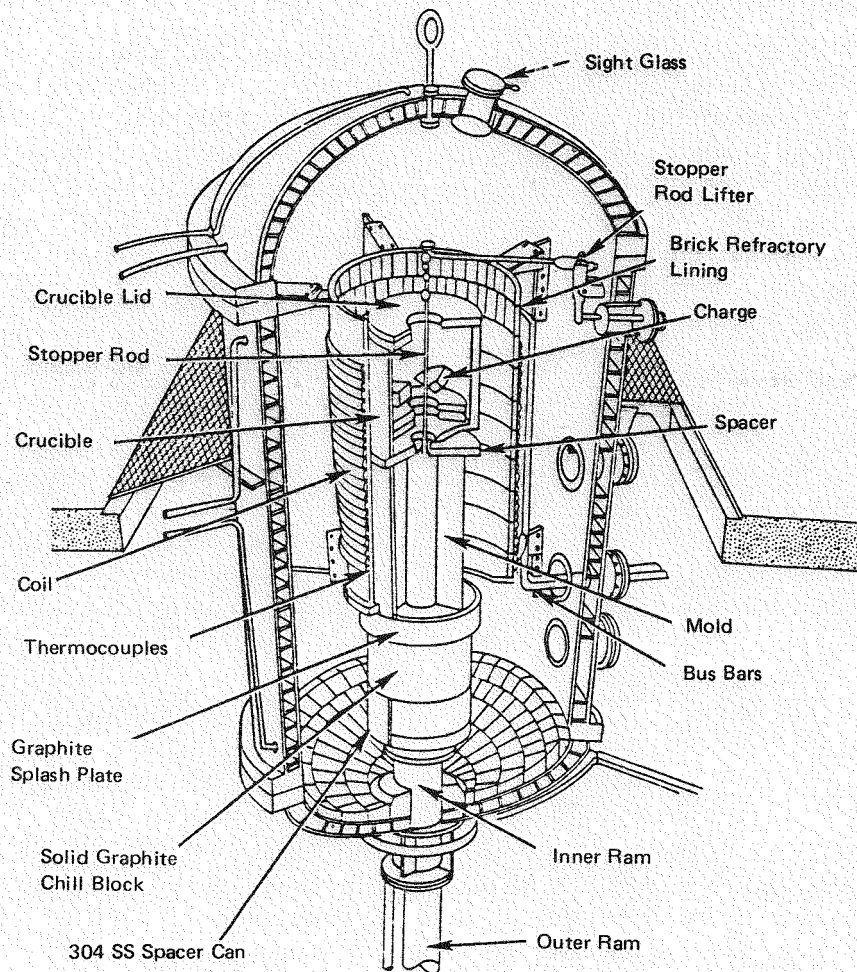
FIGURE 2. Rocky Flats VIM Furnace Showing
the Crucible and Mold in the Operating Position

firebrick and crucible-mold. Firebrick for the
28-inch coil is one inch thick, while that for the
36-inch coil is two inches thick. The furnace lining
is a special type insulating refractory material
capable of withstanding temperatures to 1525 °C.
The refractory firebrick acts as a barrier against
heat transfer from the hot crucible, mold, and
induction coil. The refractory lining is laid in place
without mortared joints with the aid of a refractory
retainer cylinder. Non-mortared joints offer two
advantages: (1) the moisture contained in the
mortar does not have to be dried, and (2) the open
joints between bricks act as expansion cushions to
accommodate severe thermal shocks during normal
operation.

The furnaces are equipped with two independent
rams within a single housing. Their functions are
as follows: The outer ram is used to lower the

entire furnace bottom for routine maintenance or
during emergencies while removing metal spills.
The inner ram is used for loading and unloading
the furnace. The furnaces are equipped with
pneumatic locks to prevent drifting of the furnace
bottom in the event of slight leakage in the ram
hydraulic system. Nonetheless, eight large "C"
clamps securely attach the furnace bottom to the
middle shell.

The furnace is top loaded; and in normal
operation, only the top needs to be opened. When
the top is opened with an overhead crane, it swings
backward and rests on a downdraft table. Between
the furnace and the downdraft table is a downdraft
grate. And, as mentioned earlier, a downdraft port
in the furnace is activated before the furnace is
opened.

Furnace Load

The inner ram mechanism raises and lowers what is referred to as the furnace load. A typical furnace load is shown positioned in the coil in Figure 2. At the bottom is a platen. The platen, attached to the top of the inner ram, is a heavy cast stainless steel plate, which acts as a base support for the spacer can. The spacer can is a stainless steel weldment specially fabricated to establish a minimum height between the inner ram and the mold. Above the spacer can is a solid graphite chill block. The chill block causes heat to flow mainly from the bottom of the mold rather than through the sides. This causes directional solidification, which is critical in uranium casting. The chill block also serves to position the crucible and mold in the coil.

The height of the chill block, spacer can combination is the preferred adjustment mechanism for positioning the mold and crucible in the induction coil. Rather than attempt to adjust the ram height, the normal operating procedure is to adjust the inner ram to the lowest position and choose a properly dimensioned spacer can and chill block to position the mold and crucible. Such an arrangement results in better reproducibility of mold and crucible positions. Furthermore, there is some question regarding possible heating of the steel ram while it is in the inductive field. Fully lowering the ram eliminates this potential hazard.

Above the chill block rests a graphite splash tray. Its purpose is to contain a spill in case a crucible or mold fails. It is critical that any spill be contained. A melt-through of the furnace bottom or wall could lead to a disastrous steam explosion. The splash tray also serves as a host shield to prevent damage of such things as thermocouple blocks and leads.

The last two major components of the furnace load are the crucible and the mold. These items are always arranged so that the molten metal from the crucible can be poured into the mold without moving the mold or crucible. To accomplish the pouring action, the crucible is placed over the mold and centered so that the bung or pouring hole in the bottom of the crucible is directly above the mold opening (sprue). The crucible is the container for melting the charge. The melting action results from inductive coupling of the coil with the crucible walls. Heat resulting from this phenomenon is then conducted from the crucible walls to the crucible bottom to the metal, subsequently melting the charge.

Above the mold rests a 1 1/4-inch-thick graphite spacer. The grooved spacer allows gas to escape when the metal is poured. Above the spacer rests the crucible, which contains the charge to be melted and cast. The crucible contains a bung and stopper rod to control the metal flow into the mold.

A controlled pouring action is maintained by placing a graphite stopper rod into the bung of the crucible. The rod is held firmly seated in the bung by the stopper rod lifting mechanism. When the metal is melted and thoroughly outgassed, the heat is poured by raising the stopper rod, thereby permitting the molten metal to flow from the crucible into the opening (sprue) directly below.

Placement of the mold within the induction coil is critical; that is to say, the mold must be positioned in the induction coil so as to establish the proper temperature gradient from the bottom of the mold to the top. A steep temperature gradient is very important to permit progressive solidification from the bottom to the top.

It is imperative to control mold temperatures as well as melt temperatures. Mold temperatures are measured by thermocouples positioned in the mold. Melt temperatures are measured by viewing through the sight glass with an optical pyrometer. Because of the crucible's placement above the mold, it is impossible to view the mold itself. For that reason, thermocouples are placed at required points on the mold. The furnace and instrument panel are capable of measuring and recording three mold temperatures: bottom, center, and top. Generally, only mold bottom and mold top are monitored.

Vacuum System

From 1953 to about 1985, mechanical roughing pumps backed by oil diffusion pumps evacuated the furnace chambers. The pumping systems were

able to evacuate the chambers to less than 50 microns with ample pumping capacity to maintain less than 100 microns during melting. Present technology suggests furnace vacuum equipment should consist of displacement and/or blower-type pumping systems rather than oil diffusion systems.

During 1985 and 1986, all the stokes mechanical and diffusion pumps were replaced with mechanical roughing pumps backed by 3000-rpm blowers. These are monoblock vacuum pumps (Leybold-Heraeus) of the rotary piston type, costing $8,500 each. The Roots blowers (Aerzen) are rated at 3000 rpm and cost $11,000 each. The blowers take over at about 150 torr. The furnace chambers are about 350 cubic feet in volume. Pumping efficiencies have about doubled with the new pumping system.

Presently, when the vacuum falls below 100 microns, the casting run may be initiated; however, it is preferable to wait until the vacuum falls below 10 microns. The vacuum increases during the casting run by 10 to 20 microns and then decreases after pour.

Power System

From 1953 to about 1985, motor generators supplied the high-frequency power to melt uranium at Rocky Flats. During that period, uranium was melted at power settings of 45 to 250 kW at frequencies of 900 to 3000 cycles per second. The four furnaces removed in the 1960s and 1980s had motor generators rated at 3000 cycles per second. The power used to melt uranium depends on the size of the casting to be made, the frequency, and the alloy content of the uranium. Mold temperature is determined by thermocouple; metal temperature is determined by optical pyrometer.

In 1985 and 1986, all the motor-generators were replaced with AC/AC solid-state SCR power supplies. The power supplies were manufactured by Ajax Magnethermic Corporation of Warren, Ohio. The units are rated at 250 kW (Model A-3) and cost $42,000 each. The power input is rated at 480 V, 340 kVA, 408 A, 3 phase, and 60 Hz. The power output is rated at 250 V, 1500 kVA,

408 A, 1000 Hz, and 250 kW. The 36-inch-diameter coils are operated at 960 Hz at a power to 275 kW. The 28-inch-diameter coils are operated at 720 Hz at a power to 180 kW.

VIM OPERATIONAL PARAMETERS

Operating procedures vary with each mold design and to some extent with identical furnaces. It is, therefore, difficult to apply a set of operating conditions that will universally produce the desired results. However, operating parameters showing the greatest influence can be examined.

Typical Casting Run

The ingot mold is first loaded and positioned in the furnace. The charge crucible is then placed directly above the mold, almost touching it and carefully aligned with respect to the mold. Insulation is placed around the crucible and mold as needed. Generally, the upper part of the mold is insulated and the bottom is not. The crucible/mold is lowered through the coil and positioned such that the top part of the mold is slightly (two to three inches) in the coil. The mold then receives heat from both the heated crucible above it and the induction coil. This heating, combined with the insulating sleeve around the upper part of the mold, creates a steep temperature gradient from top to bottom. When the melt is poured, the temperature gradient causes directional solidification to proceed from the bottom to the top of the ingot. This casting practice normally produces ingots with no internal cavities. An improper solidification pattern leads to axial cavities and trapped impurities, which is a nonacceptable condition. Another important benefit derived from directional solidification is that objectionable impurities concentrate in a shallow top layer of the ingot, which can be cropped.

After the furnace is loaded and sealed, it is evacuated by vacuum pumping. When the pressure drops to 30 microns, the water is turned on and checked for proper flow. After the pressure drops to 10-20 microns, induction heating is started. For a large uranium charge, the power is slowly increased to 100 kW. After 15 minutes at 100 kW,

the power is increased to 180 kW, held 15 minutes, and then increased to 220 kW. Pumping and heating are continued until the melt temperature reaches about 1425 °C; the temperature is then dropped (by reducing power) to 1350 °C and the melt is allowed to outgas for about 30-45 minutes. The melt is not poured until the charge stops bubbling. Magnesium, slag from reduction oxidation products, hydrogen, and radioactive decay products such as thorium and radon float to the top of the melt. The volatile impurities, of course, distill from the melt. Before pouring, the mold bottom temperature is checked to ensure that it is above 700 °C and below 850 °C. The power is then turned off, the furnace is isolated from the pumping system, and the pour is made manually by actuating the pouring mechanism, which lifts the coated graphite bungs. The pour is complete after about two minutes. When the ingot cools to about 300 °C (a temperature at which uranium does not oxidize excessively), the furnace is brought to atmospheric pressure with nitrogen and opened. The overall time for a heat is 11 hours, with pumpdown and heatup requiring three hours, melting and casting one hour, cooldown six hours, and loading and unloading another hour. This cycle can be reduced about three hours by admitting helium gas to the furnace chamber shortly after pouring to hasten cooling.

Uranium Melting and Casting Characteristics

Uranium has a relatively high melting point (1131 °C), high boiling point (~3820 °C), high heat of fusion (20 cal/g), and high heat of vaporization (~450 cal/g). The large liquid range (over 2000 °C) and the accompanying low vapor pressure in the normal melting and casting temperature zone (1300-1450 °C) are very desirable. One difficulty in melting and casting of uranium is the temperature required to maintain fluidity during castings, i.e., 1300-1450 °C. Not only are these temperatures costly to achieve, but more importantly, uranium at these temperatures is a highly reactive metal. It is highly reactive with most container materials and most atmospheres.

A vacuum or truly inert atmosphere is required for melting and casting uranium. Also required is a strong container material that can be made inert to liquid uranium. A good heat source is required to keep things hot. Fortunately, vacuum-induction

melting and coated graphite crucibles are well established technologies, thus allowing easy melting of uranium and a high-purity product in the form of defect free castings when done properly.

Another factor in melting and casting uranium is its high density in the liquid state (16.6 at 1140 °C). This requires a strong container material capable of withstanding high loads as well as high mechanical and thermal shock. Foundry buildings actually shake when a heat of uranium is poured. But the high density and corresponding high hydrostatic head do aid cavity filling, thus producing a sound casting.

Virgin Metal Feed

The virgin or source metal for VIM refining and casting is depleted derby uranium and scrap. Depleted uranium is a by-product of uranium enrichment plants. It contains less than 0.71% of the fissionable U-235 isotope. Naturally occurring uranium contains 0.71% U-235, while enriched uranium contains more than 0.71% U-235. Depleted uranium produced in the United States normally contains 0.2% U-235. The casting concepts outlined in this report apply equally to natural, enriched, and depleted uranium, except that nuclear criticality concerns limit the charge and ingot size for normal and enriched uranium, especially for highly enriched uranium.

Depleted derby uranium is the end product of reducing the depleted UF_6 by-product of the uranium enrichment facilities. This is a two-step reduction: 6-4 and 4-0 reductions.

The source of all virgin metal for melting and casting at the Rocky Flats Plant is the Feed Metal Production Center at Fernald, Ohio operated for the DOE by Westinghouse, formerly operated by National Lead of Ohio (NLO).

Uranium metal is produced at Fernald by reducing uranium tetrafluoride, UF_4 (green salt), with magnesium metal flakes. The chemical reaction is represented by the following equation:

$$UF_4 + 2Mg = U + 2MgF_2$$

$$(\Delta H^{\circ}_{298 \, ^{\circ}C} = -83.5 \text{ kcal})$$
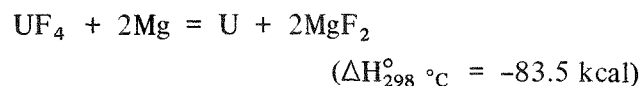
A small excess of magnesium is required to achieve maximum yields. This thermite-style reduction of

uranium tetrafluoride is carried out in a closed steel vessel insulated with a refractory lining of the reduction by-product, magnesium fluoride. The sealed steel reaction vessel is placed within a gas-fired furnace and the mixture self-ignites at about 600 °C. Sufficient heat is generated in this exothermic reaction to allow near-complete gravity separation of the light molten magnesium fluoride and the dense molten uranium metal. After cooling, the regulus of solidified metal at the bottom of the vessel (uranium derby) is removed.

The Fernald process described in the previous paragraph produces a derby with a nominal "as reduced" weight of about 160 kg, measuring about 4.5 inches in height by about 12 inches in diameter. However, the techniques for reducing uranium tetrafluoride are basically the same whether the product is a 25- or a 1500-kg derby. Typical parts-per-million impurity levels of derbies melted and cast at the Rocky Flats Plant are: aluminum 8-40, carbon 10-50, copper 5-50, hydrogen 2-10, iron 30-100, magnesium 1-10, nickel 10-50, nitrogen 8-40, and silicon 10-80.

The derby from the reduction operation must be cleaned and then melted and cast under vacuum. Vacuum melting reduces the level of certain objectionable impurities and permits casting of the metals into ingot form.

Graphite

Graphite is currently used exclusively in uranium VIM foundry casting operations because it is readily available, is easy to machine, has good thermal-shock resistance and heat-transfer properties, is relatively inexpensive, and is some-what inert, i.e., not highly reactive to molten uranium. When protected by a painted ceramic coating, the graphite components are reusable and more economical than any known ceramic substitute.

Crucibles, molds, and other graphite components for vacuum-induction melting and casting of uranium are machined from high-grade graphite blocks and rounds. The graphite stock must meet an HLM or equivalent specification. The major suppliers of graphite stock to Rocky Flats are

Great Lakes Carbon Corporation and Union Carbide Corporation.

Stick graphite is the base material from which crucible/mold components are machined. Sticks 24 inches or less in diameter are normally made by the extrusion process, varying in grade and physical properties. Sticks 36 inches in diameter are made by molding. Experience shows that as the diameter increases, there is increased porosity and decreased strength. This is especially true for graphite sticks more than 12 to 14 inches in diameter. Grades of graphite materials such as CBB or H4LM are preferable for crucibles; CS312 and ATL are preferable for molds. Great Lakes Carbon Company makes H4LM, while CBB, CS312, and ATL are products of National Carbon Company.

Manufacturers will furnish nominal physical properties for grades of graphite they produce. These data can be used in the design of crucible/mold parts with some assurance of non-failure. For general requirements, graphite having a density above 1.60 g/cm^3 is desirable. Properties may vary from stick to stick within a given grade. It follows that material selected for critical use should be carefully examined for quality to avoid failure of the part. When a question arises regarding strength of a particular stick, it may be wise to select samples for mechanical tests.

Crucible Design

At the Rocky Flats Plant, production VIM foundry graphite crucibles of two widths are normally used: a 35-inch-diameter crucible for the 42-inch diameter inductance coil and a 23-inch-diameter crucible for the 28-inch coil. The 35-inch-diameter crucible is machined from 38-inch-diameter graphite stick, while the 23-inch crucible is machined from 26-inch diameter stick.

A representative 35-inch-diameter crucible is dimensioned in Figure 3. The crucible shown has three pour holes for casting three separate ingots in one casting run. If only one ingot were to be cast per run, the pour hole would be in the center of the crucible. Each pour hole is a replaceable bung. Each bung is 2.5 inches in diameter, 1.0
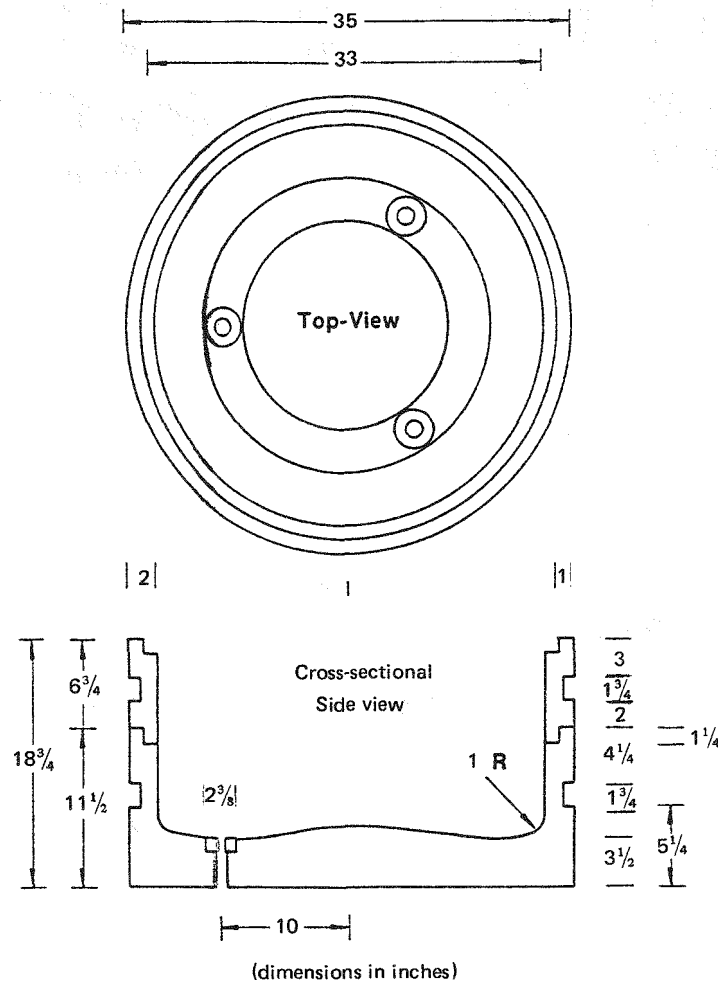
FIGURE 3. Representative 35-Inch-Diameter Graphite Crucible With Three Pour Holes for Gang Casting Three Cylindrical Ingots in One Run

inch thick, and has a 7/8-inch-diameter opening with a 5/8-inch radius on the top. The 1-1/4-inch diameter stopper rod also has a 5/8-inch matching radius on one end. The grooves on the side of the crucible are for lifting. The grooves on top of the crucible are to support the graphite lid. The crucible lid is 1-1/4-inch-thick graphite and has three five-inch-diameter openings to accept the three stopper rods. All crucibles are designed with the bottom tapered three to five degrees toward the pouring holes. This construction minimizes the skull (metal + oxide) hangup in the crucible.

A 23-inch-diameter crucible is similar in design; however, it has only one pour hole since there is not room for the charge plus additional stopper rods. Also, the 23-inch-diameter crucibles are generally considerably higher to accommodate a reasonably sized charge.

If a crucible makes it through the first run, it generally lasts 100 to 200 casting runs. The cost of an average 35-inch-diameter crucible is about $3,600. Bungs generally last through 20 to 30 casting runs and are resized after each run. Stopper rods last about 20 to 50 casting runs and are resized after each run.

Mold Design

Split graphite molds are used extensively at the Rocky Flats Plant to vacuum-induction cast

uranium. Major advantages of a split mold compared with a solid or near-solid mold are: (1) ease of casting removal, (2) mold tapers not required for ingot removal, (3) economy of replacing a damaged mold segment rather than the whole mold, (4) reduced chance of mold damage on casting removal, and (5) ease of machining elongated cavities. Major disadvantages of a split mold are: (1) protruding seams in the ingot that mirror the mold junctions, and (2) increased mold erosion and damage at the mold junctions. For most rolling and extrusion ingot designs, split molds are more economical than monolithic molds.

Molds must be designed to compensate for shrinkage of the casting during solid-state cooling. Molds are dimensioned to provide for 1.8% shrinkage on the diameter and 2.2% on the length. Where castings are to be machined, 1/8 to 1/4 inch is generally added to the dimension to allow for surface cleanup. The dimensional amount added frequently depends upon the size and shape of the casting and the requirement of the machine shop. Satisfactory uranium castings have been made where only 0.050 inch was allowed on a surface for machining operations.

A split mold used to cast 4-inch-thick rolling ingots is shown in Figure 4. The replaceable insert increases the life of the mold bottom significantly. The mold is dimensioned in Figure 5. Mold bottoms and tops of this type have a life of 30 to 40 casting runs. Mold inserts have a life of 10 to 20 runs. The cost of this type mold is about $3500.

A typical split mold used to cast extrusion or rod-rolling ingots is shown in Figure 6. At the outer middle of the side molds is a half-inch protrusion tapering toward the mold top over a distance of 4 3/4 inches. This taper is mirrored on the inside of the belly ring to fix its position on the assembled mold. The L-groove at the top outside of the side molds is to position the top ring. The U-groove between the taper and the L-groove is for lifting. The L-groove at the bottom outside of the side molds is to form a better seal in the assembled mold.

The top ring and belly ring have lives of 100 to 200 casting runs; the mold bottoms last 40 to 50 runs,
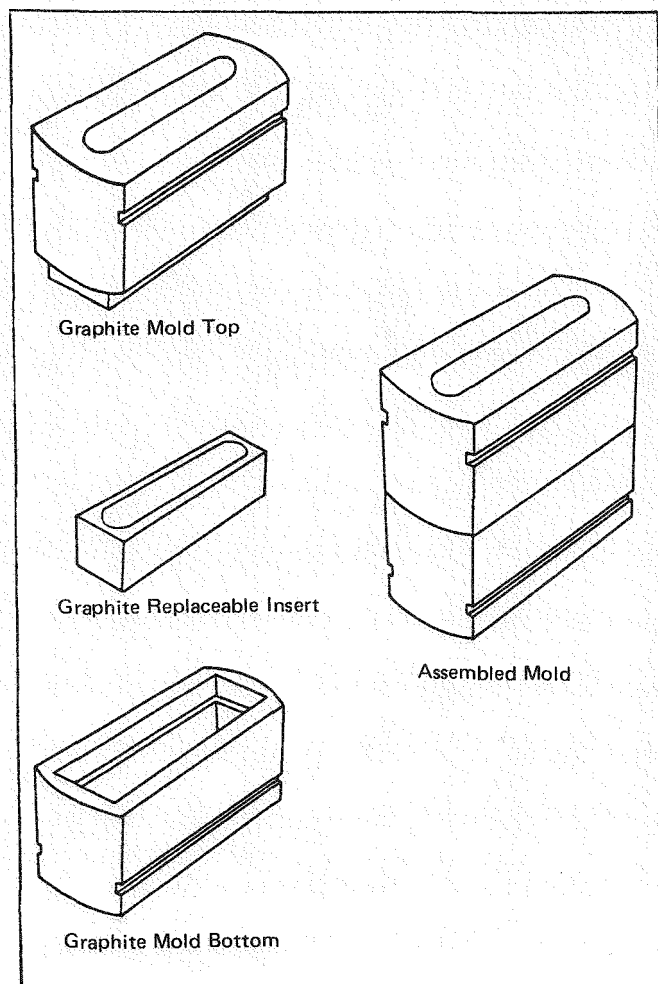
FIGURE 4. Split Graphite Mold Used to Cast a Four-Inch-Thick Rolling Ingot

while the mold sides last 15 to 25 runs. Foundry molds of this type have inside dimensions producing ingots of 4, 5, 6, 8, 10, 12, and 14 inches in diameter. Heights of the mold vary from 36 inches for the small diameters to 24 inches for the larger diameters. Costs vary from $1000 to $1600.

For casting hollow shapes, a graphite core is used. For straight hollow cylinders, the core is generally not reusable, since machining is required for removal. To hold mold components together during casting, a press fit between the case and core plus graphite pins for added strength to the assembly has proved satisfactory. Press fits vary in relationship to diameter and can be grouped as follows: for fitted diameters 0 to 10 inches, use 2- to 3-mil interference; for fitted diameters 11 to 18 inches,
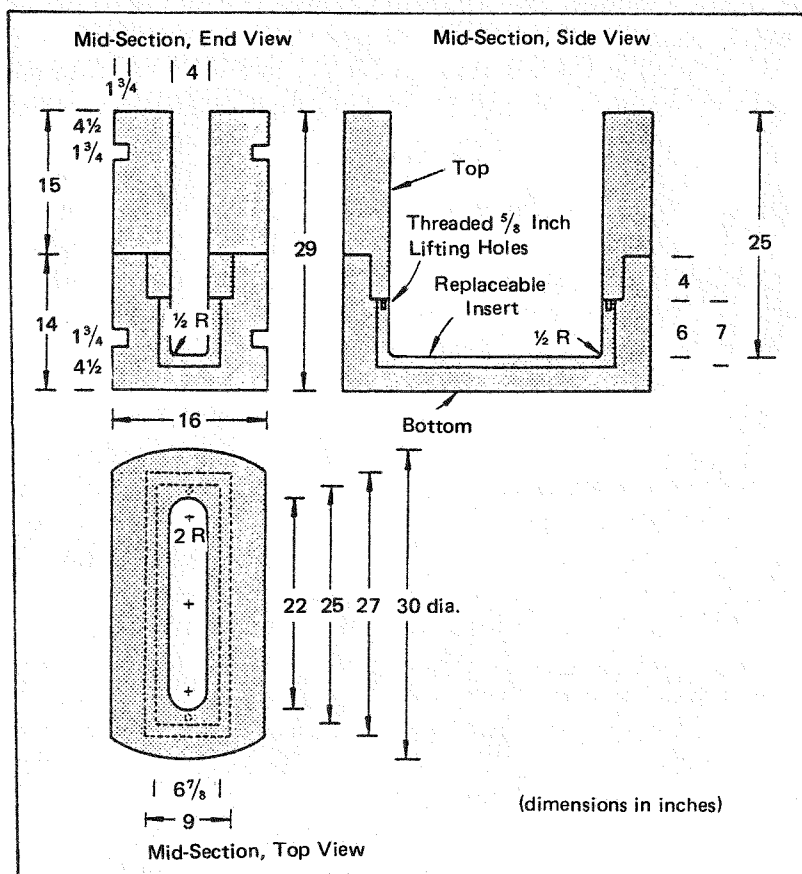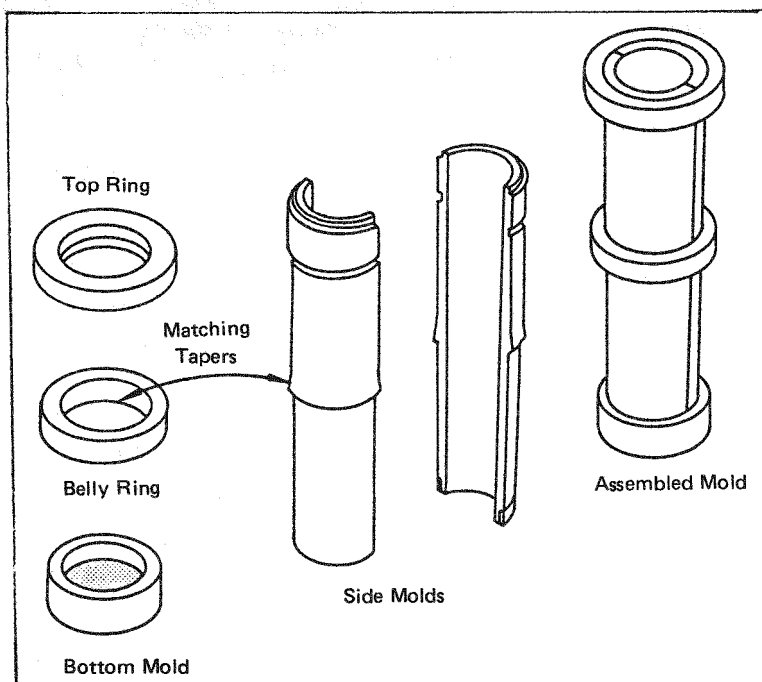
FIGURE 5. Split Graphite Mold Used to Cast a
Four-Inch-Thick Rolling Ingot. (dimensional views)

FIGURE 6. Split Graphite Mold Used to Cast
a Cylindrical Extrusion or Rod-Rolling Ingot

use 3- to 4-mil interference; for fitted diameters 19 to 24 inches, use 4- to 5-mil interference; and for fitted diameters 25 to 38 inches, use 6- to 7-mil interference.

Graphite pins are designed according to the buoyant forces exerted by the charge weight and volume displacement. Experience shows that the use of push fits for graphite pins is better than interference fits. Because graphite expands rapidly as the temperature increases, the stresses set up by excessive interference fits can easily cause cracking.

For intricate or thin-walled castings, a hot-top or riser can be built into the mold for feeding purposes. For most purposes, a hot-top or riser containing 20 to 25% of the casting volume provides an adequate molten metal reservoir to feed the casting during solidification. Consideration must be given to placement and dimensions of this hot-top to secure optimum feeding.

Two properties of uranium that also aid in securing sound castings in metal are fluidity and high density. Both characteristics contribute to filling a mold completely and maintaining feeding as solidification takes place.

Refractory Coatings

A refractory coating on the graphite that contacts molten uranium is mandatory for high-purity, high-quality castings. The main purpose of the coating is to prevent metal-graphite reaction, thereby reducing carbon pickup in the metal. Unalloyed uranium melted and cast in uncoated graphite crucibles and molds pick up about 400 ppm of carbon. Certain uranium alloys, such as U-Nb and U-Zr, when cast into uncoated graphite molds form a finely-divided, carbide-loaded surface that can explode during or after immersion in nitric acid pickling.

Refractory coatings are absolutely required when melting uranium alloyed with carbide-forming elements. The carbide-forming alloying elements react with carbon in the uranium as well as with exposed carbon of the crucible to form solid carbides. The carbides formed have relatively low

densities and tend to float upward in the melt (liquation) or in the casting before and during solidification. Such behavior results in drossing out the alloying element and/or pronounced segregation of the added element. A coating on the crucible and mold that effectively prevents metal-graphite reactions also prevents carbon pickup in the uranium melt and therefore reduces element loss through carbide formation and liquation.

Refractory crucible/mold coatings for uranium are usually based on finely divided refractory oxides and zirconates. Ideally, a coating should be mechanically stable, resistant to thermal shock, inert to liquid uranium, and harmless as a contaminant. Water slurrys of stabilized zirconia or magnesium zirconate are common mold washes for graphite crucibles used for uranium. Experience shows that a water slurry of yttria works best for molten uranium metal. The mixing and application of this crucible/mold wash is discussed in this report.

The yttria crucible/mold wash was developed[85-88,99] at the USDOE Y-12 Plant at Oak Ridge, TN. Jessen[114] compares yttria against other popular coatings based on the average carbon increase in uranium as a function of the run temperature and crucible coating type. The results are summarized in Figure 7.

Mold washes for melting uranium in graphite may be applied by brush, roller, swab, or spraying. The viscosity should be controlled to a consistency suited to the mode of application. A layer of at least 0.010 inch is normally desirable to prevent metal penetration. To consolidate the coating, the liquid medium must be evaporated. Water-based coatings may be dried by air, gas torch, or infrared heating. A coating must dry without cracking or peeling to provide a hard, tough layer. Binders are frequently added to the wash to provide this function. Coating thickness greater than 0.015 inch are usually avoided because of their tendency to spall.

Storage, application, and drying are important when using a liquid wash. To delay settling of the solids while standing, all constituents should be in a fine state of division. Settling is further restricted by suspending agents. A uniform paint consistency
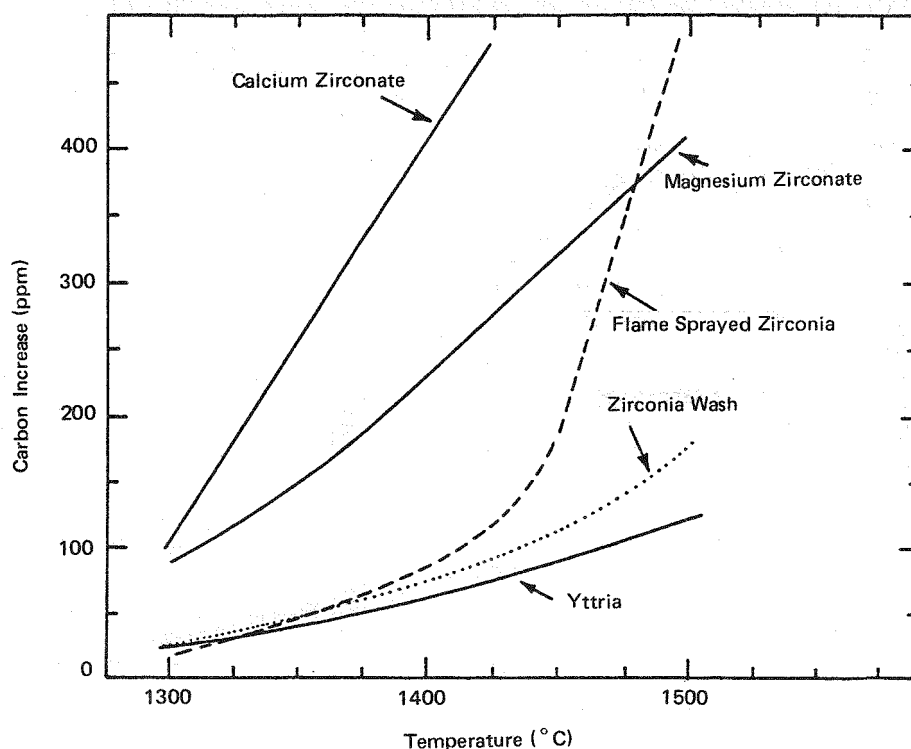
FIGURE 7.   Average Carbon Increase as a Function of the
Run Temperature and Crucible Coating Type. (Reference 114)

can be assisted by continuous agitation in the container, usually by mechanical stirring.

Ceramic coatings may also be applied to the exterior surface of molds to reduce heat losses. The ceramic, being a poor heat conductor as well as a thermal reflector, tends to retain heat in the mold. By coating the upper exterior surfaces of a mold, heat losses are reduced, thus promoting improved thermal gradients with better directional solidification. This practice can be applied to long molds with lower portions outside the effective heating range of the induction coil. By coating exterior mold surfaces on the lower and cooler parts, heat losses are reduced.

Mold washes can also help metal fluidity by reducing frictional forces dependent on the roughness of the mold surface. This may be because of the reduced friction of a smooth surface or the altered wetting characteristics of the metal-mold interface. Mold washes have also been used to inoculate a melt to provide finer grain size in castings other than uranium castings.

In summary, experience shows that yttria is the best all-around coating for melting and casting uranium in graphite crucibles and molds. Yttria works better or as well as stabilized zirconia or magnesium zirconate in preventing reaction between uranium and graphite, and its real beauty is its ease of stripability from crucibles and molds. Yttria is highly recommended for both crucibles and molds. It is used exclusively at the Rocky Flats Plant.

Furnace Stack

Other necessary components in the furnace stack are made of graphite. A typical furnace stack is shown in Figure 8. The stack shows a 35-inch-diameter, center-pour crucible positioned to cast a four-inch-thick rolling ingot. The reason for using a solid graphite chill block rather than a stack of fire brick is to take advantage of the relatively high heat transfer property of graphite. Taking heat preferentially from the bottom of the molds rather than the sides promotes directional
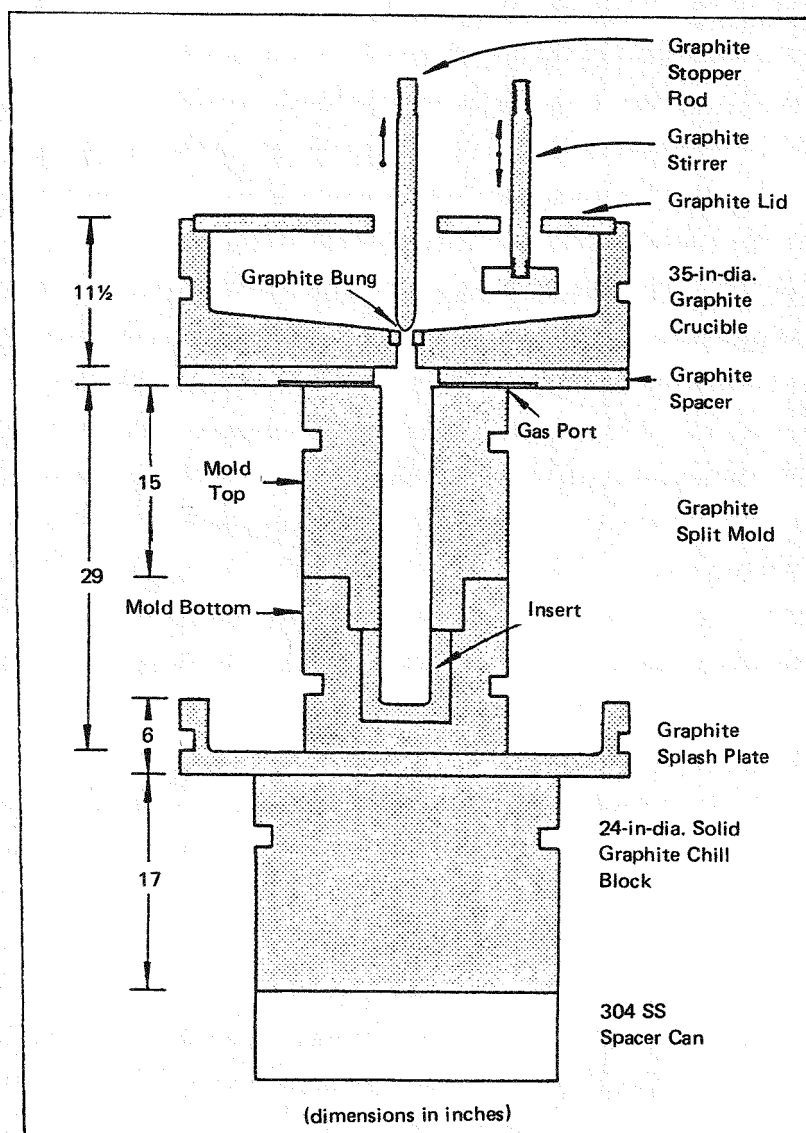
FIGURE 8. Typical Furnace Stack Showing a 35-Inch-Diameter Center-Pour Crucible in Position to Cast a 600-kg, 4-Inch-Thick Rolling Ingot

solidification, which is desirable. Chill blocks are available in several heights as are the underlying spacer cans. Combining the two helps fix the overall stack height. Molds vary in height; and by adjusting the pedestal height (chill block plus spacer can height), the molds and crucible can be firmly supported in a selected position in the furnace. This is necessary to obtain the proper temperature gradient in the mold in a single-coil furnace.

A 35-inch-diameter graphite splash dish centered on the chill block acts as a reservoir to contain molten metal in case of a spill or mold breakage during casting. The spilled metal flows into the well where it solidifies. Generally, this metal needs only slight cleaning to be used in another charge; however, certain uranium alloys when cast directly on graphite form a surface that can be explosive when cleaned in strong oxidizing acids such as nitric acid.

Between the mold and crucible is a graphite spacer. In this case the spacer is 35 inches in diameter, 1 1/4 inch thick, with a 5-inch-diameter hole at the center. On the bottom of the spacer are four U-grooves 90 degrees to each other, radiating from the center hole. The grooves are 1 inch wide and 1/4 inch in depth. The main purpose of the grooves is to allow gas to escape during pouring. They also serve as ports in pumpdown of the mold cavity. In gang castings, larger openings are desirable in case a stopper rod breaks and fails to lift. The larger openings would be necessary to accommodate metal overflow in some of the molds. For gang castings, three graphite pucks (2-inch diameter by 1 inch thick) are placed between each mold and the spacer plate. The spacer plate used in gang casting is not grooved. When three cylindrical ingots are simultaneously cast, the spacer plate has three holes 120° to each other to allow molten metal to pass from the crucible into the molds.

A graphite lid sits in an L-groove on the top interior of the crucible. The lid is 32 inches in diameter, 1 1/2 inches thick, and has openings for the stopper rods and a stirrer. The graphite stopper rod is 1 1/4 inch in diameter and can vary in length depending on the overall stack height. One end of the stopper rod is threaded for attachment to the lifting mechanism. The other end is hemispherical for proper seating in the bung. The graphite stirrer is a plunger type. The head is six inches in diameter and two inches thick with a centrally located threaded hole on one side. The threaded hole accepts a threaded 1-1/4-inch-diameter graphite rod.

The furnace stack shown in Figure 8 is not complete in that carbon felt insulation is normally part of the furnace stack. Generally, the crucible needs additional insulation and the molds do not. Carbon felt is normally placed between the spacer and crucible and above the graphite lid. The crucible is also surrounded with a layer of carbon felt.

Crucible Charge and Metal Yields

The crucible charge for melting can be virgin metal (derbies), scrap recycle, or any blend of the two. Charge weights at the Rocky Flats Plant are normally about 450 to 650 kg. The largest casting ever made at Rocky Flats was a 17-inch-diameter, 24-inch-long extrusion ingot weighing 1500 kg. This is the approximate safe upper limit because of the 250-kW power supply and overhead cranes with two-ton lifting limit at Rocky Flats. A common derby-scrap charge blend would consist of two derbies at 160 kg each plus scrap to achieve a total charge weight of 480 kg. No effort is made to clean the scrap prior to placing it in the crucible. Large chunks of metal require considerably longer times to melt than equivalent weights of smaller chunks. So melting times are dependent on size distribution of the charge.

Virgin metal charges normally yield 97–98% cast metal. Charges containing all solid scrap (no hot-tops) normally yield 95–96% cast metal. Charges containing all briquetted turnings would be expected to yield 75–80% cast metal. Turnings are generally blended at the 15% level to maintain a healthy stream chemistry. At the present time, turnings are not recycled at Rocky Flats. At Rocky Flats, the charge composition is recorded for posterity. Ingots of all virgin metal are lettered differently (160XXX) from those containing scrap (402XXX).

A word of caution is in order about loading crucibles with uranium metal. Considerable care must be taken not to damage the thin mold wash or the underlying graphite. Uranium metal is very heavy. This, coupled with the many sharp corners present, will readily remove the crucible wash unless care is observed. Scrap charges are hand loaded.

The purity of a casting depends on the purity of the charge. As a rule of thumb, a melting and casting cycle increases carbon 20 to 40 ppm and iron and silicon 5 to 10 ppm. Thus, certain impurities build up in recycle streams with time, and blending with virgin metal is required. Generally, a decent chemistry can be maintained with a 50-50 virgin-recycle charge.

Gang Casting

In the vacuum-induction melting and casting of rolling and extrusion ingots, it is common technology to cast several ingots in one heat. This

casting of multiple ingots is referred to as gang casting.

A typical crucible-mold design to produce three rolling slabs in one heat is shown in Figure 9. The mold is a split design using a cope and drag approach. The mold has three cavities, each with a one-inch radius on the sides and bottom. The mold produces three ingots, measuring 16 inches wide by 20 inches high by two inches thick, each weighing about 175 kg. The crucible has three bungs (not shown), which are positioned over the center of each cavity. Three graphite stopper rods are seated in the bungs and are attached to a common lifter.

A mold that produces the same type ingot one at a time is shown in Figure 10. Ingots produced in the single-cavity or triple-cavity mold appear identical, and both molds produce excellent castings. The triple cavity mold is preferred because of its higher throughput. The average triple-cavity mold life is 10 heats or about 30 ingots. This compares to a single-cavity mold life of 30 heats and 30 ingots. The advantage of the triple-cavity mold is being able to gang-cast 30 ingots in 10 working days compared with 30 single-cast ingots in 30 days. Evidently, one working day is required per heat.

Bottom Pouring Versus Top Pouring

In uranium production vacuum-induction melting and casting furnaces, bottom pouring is the standard procedure. The reasons are: (1) simplicity and economics of furnace design, (2) simplicity of operation, (3) less chance of slag entrapment in ingots, and (4) the crucible and mold can be heated with one induction coil.

FIGURE 9. Typical Crucible-Mold Design to Produce Three Rolling Ingots in One Heat
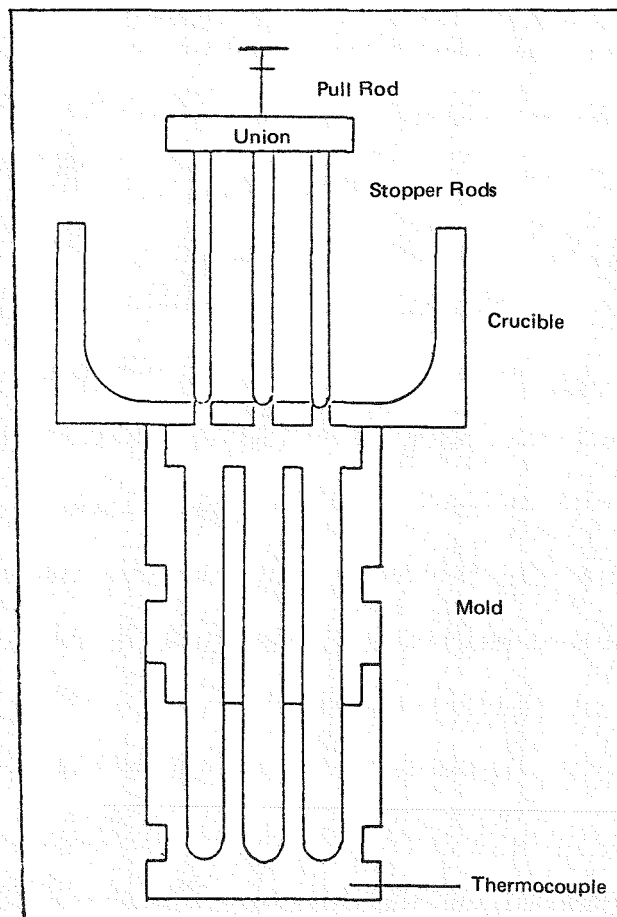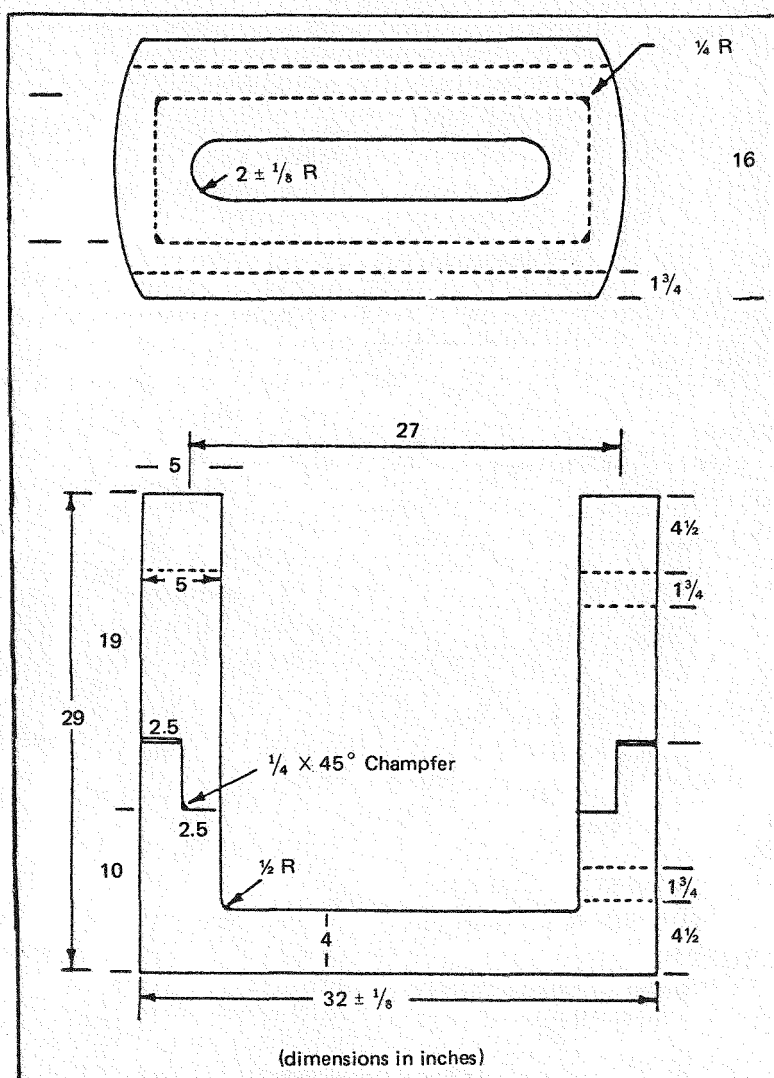
FIGURE 10. Single-Cavity Split
Mold That Produces the Same
Ingot Size Gang Cast in Figure 9

A major disadvantage of bottom pouring is that the metal is initially at a high metallostatic (hydrostatic) head. The metal is apt to stream out rapidly and uncontrollably, and the incidence of mold erosion and cold shuts is greater. In top-pour furnaces, the "rocking chair" principle permits accurate control of the metal stream entering the furnaces, and mold erosion and metal splattering can be more closely controlled.

Another disadvantage of bottom pouring is the chance of bung or plug failure or leakage resulting in the premature entry of metal into the mold and defective castings. Another disadvantage of bottom pouring when using a plug is the chance that the plug may become entrapped in a usable part of the casting rather than in the hot-top. Even when the plug properly floats to the top of the metal, the hot-top is less usable or recyclable with an entrapped plug.

Modern vacuum-induction, tilt-pour furnaces have vacuum-sealed manipulators for removing slag prior to pouring, as well as for breaking bridged metal and for immersing dip thermocouples. Some of these furnaces are designed so that pouring and charging

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can be done semi-continuously while the vacuum and the furnace temperature remain constant. Versatile furnaces provide for mold changes so that casting dimensions can be readily changed. Many uranium melting operations are carried out so meticulously and the charge is so pure that slag content is low enough to use top-pour furnaces without skimmers. However, this remains to be proven on a production scale.

An advantage of top pouring is in homogeneity of the casting. During melting, there is a tendency for lighter elements to segregate upward. In top pouring, the top of the melt becomes the bottom part of the casting. In proper casting, the ingot remains molten for a short period and solidifies from the bottom up. This allows a chemical mixing or homogenization that is not possible in a bottom-pour furnace. In defense of bottom pouring, the turbulence caused by bottom pouring into a hot mold may be enough to effect a chemical mixing or homogenization equivalent to that of top pouring. This indeed holds true for the U-3/4 wt% Ti alloy using certain mold sizes.

Very little effort has been expanded in the United States for evaluating top-pour furnaces for uranium castings. According to Schimmelbusch,[116] very reliable vacuum-induction tilting furnaces are commercially available in Germany and are predominantly used there for melting uranium. Bottom-pour furnaces are viewed as rather primitive by some advocates of vacuum-induction tilting furnaces, some of whose arguments are presented above. Nonetheless, bottom pouring has been the standard technology for vacuum-induction melting and casting of uranium for more than 40 years; it does an admirable, economical job, and it will probably remain the standard technology for the next 40 years.

Pour Rates

Pour rates are a concern in controlling the quality of an ingot. The rate should be such that the ingot molds fill rapidly and uniformly without excessive splashing, turbulence, and mold erosion. Splashing, turbulence, and mold erosion are controlled by the pour rate, which in turn is controlled by the diameter

and geometry of the pour hole, the hydrostatic head, the distance of fall, and the fluidity or temperature of the metal. At Rocky Flats a pour-hole diameter of 7/8 inch is used for all routine operations. This size works well for most crucible-mold designs and most uranium alloys.

The stream of metal that first falls onto the floor of the mold may cause excessive erosion. This effect increases as the temperature of the molten metal increases and with the distance the metal falls. Also, the effect increases with the height of the hydrostatic head and inversely to the diameter of the opening. The breaking away of small chunks of graphite probably does not increase carbon content significantly, since most of the chunks or granules immediately become entrapped by solidified metal at the interface or float to the top of the molten ingots. Entrapment of the graphite chunks or granules within the ingot would, of course, cause a major or minor defect, depending on the size of the graphite. Observable graphite within the ingot from mold erosion is a very rare occurrence. However, it is possible that frequently observed small congregations of uranium and alloy metal carbide inclusions are due to very small graphite granules being entrapped and dissolved in the molten metal and later precipitated as carbides.

The loss of molds is mostly due to erosion, and erosion is most common where molten metal first contacts the mold. Replaceable graphite inserts have been used routinely and successfully at the Rocky Flats Plant to extend mold life. Using molds split horizontally allows replacement of the eroded bottom portion to reduce overall mold replacement costs. They are used routinely at Rocky Flats.

Metal splashing and turbulence are of concern in controlling the quality of an ingot. Splashing and turbulence increase as the temperature of the molten metal increases and with the distance the metal falls. Also, the effect increases with the height of the hydrostatic head and inversely to opening diameter. Splashing of metal during pouring can lead to cold shuts in the ingot and erosion on the sides of the mold. Erosion is due to high-density, moderate velocity droplets or streams of metal impinging on the mold as pouring progresses. Mold erosion of this type is pronounced where impingement is at a seam or defect in the mold. Cold shuts

are due to droplets sticking to the mold walls or bottoms and not being redissolved by the advancing molten metal. Properly cast uranium and uranium alloy ingots do not contain cold shuts.

Metal turbulence during pouring is both good and bad. Turbulence causes mold erosion as high-density metal sloshes in the mold. Another disadvantage is that turbulence can cause some of the skull or dross from the top of the crucible melt to become entrapped in the usable part of the cast ingot. Fortunately, in a proper directionally solidified ingot, the mixed-in dross has time to float from the ingot. On the pro side, turbulence can erase segregation of alloying elements and impurities that exist in the crucible melt at the time of pouring. Metal turbulence can cause considerable mixing during pouring.

Graman and Kettler[140] at the Feed Metal Production Center (Fernald, OH) conducted a study to determine the effect of orifice configuration on the integrity of a pour stream simulating molten uranium. The effect of the mold's bottom shape on the initial metal splash was also investigated. The apparatus used consisted of a 3/8-scale acrylic plastic model of the bottom-pouring crucible and mold used in production. Mercury was used as the fluid in these studies

because many of its physical properties are similar to those of molten uranium. The changes in the pour stream and in the initial metal splash were recorded with a motion picture camera. A pour plug having an orifice with tapered sidewalls and a rounded edge at the top produced the most desirable pour stream. Mold bottoms that were flat or approached flatness with no abrupt change in contour minimized metal splash.

The pour rate is proportional to the diameter of the pour or drain hole. Jaynes[75] et al. determined this relation for uranium, and their results are shown in Figure 11. Cadden[76] et al. report that Sokurski[119] et al. have determined a similar curve. Cadden et al. also report that a pour rate of 400 kg/min for a U-0.75 wt% Ti alloy heated to 1350 °C produces good ingot surfaces with no apparent mold erosion.

Melt Superheat

In most cases, it is advantageous to melt at the lowest temperature and shortest hold time possible. This is mainly due to deterioration of the crucible coatings as temperatures and times increase and, to lesser extent, the reaction of the melt with the

FIGURE 11. Pour Rate as a Function of Pour Hole Diameters for Uranium. (Reference 75)
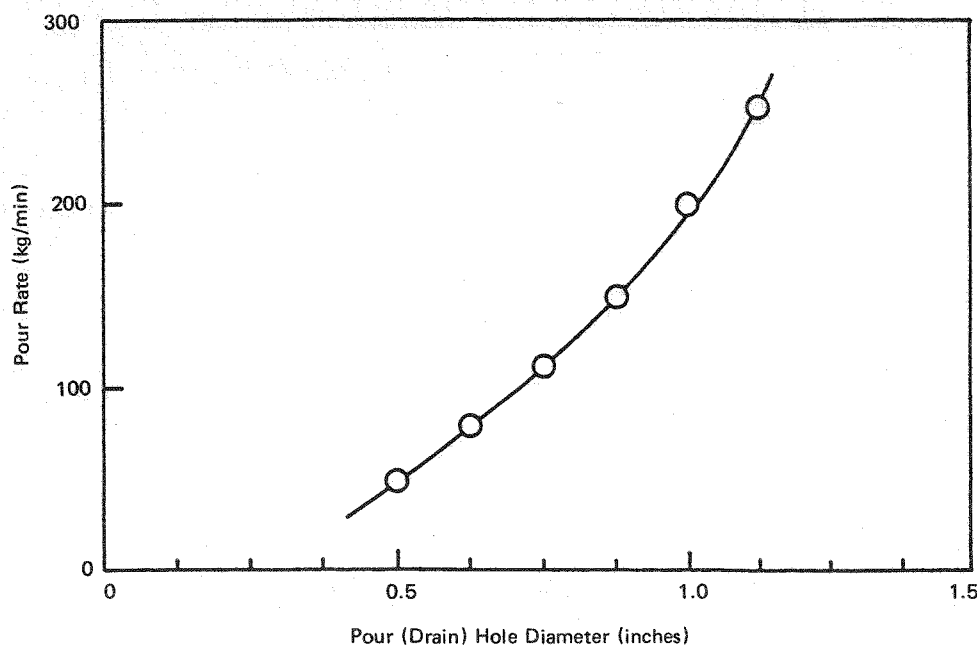
furnace atmosphere. Excessive pour temperatures can also cause problems in the mold, resulting in ingot sticking and mold damage.

The melting point of uranium metal is 1131 °C. To reduce the possibility of cold shutting in the casting and to increase fluidity of the molten metal, the pouring temperature chosen is 1330 ± 10 °C. This range is used when mold temperature gradients are from 700 °C at the bottom to 1100 °C at the top of the mold. In cases where a mold bottom temperature cannot be achieved, a pouring temperature of 1350 °C or slightly above is used. Pouring temperatures below 1300 °C frequently cause cold shutting. This is especially true for thin-wall castings. In casting a uranium alloy, the pouring temperature selected is 200 °C above the liquidus temperature. However, temperatures above 1450 °C are rarely used, and lower superheats may be used for very high melting range alloys. If the superheat is not sufficient and the mold is properly heated, cold shuts will be obvious on the cast alloy.

In general, the shapes into which uranium is cast are relatively simple. No complicated gating nor risering problems exist. This condition, together with the fact that uranium at its pouring temperature is a highly fluid metal, simplifies the transfer of the charge from the crucible into the mold. In most uranium casting operations, the metal flows from the crucible pour hole directly into the mold cavity or into a runner box that channels the flow into the mold. The rate at which the mold fills is dependent on the pour hole diameter.

Melt Temperature Gradients

Temperature gradients in the crucible melt can cause impurity and possible alloy segregation effects. Work[114] at the USDOE Y-12 Plant shows that temperature gradients can indeed exist in uranium melts. Figure 12 shows temperature profiles and, hence, poor mixing conditions in a

FIGURE 12. Thermal Gradients in a Crucible Containing a 10-Inch-Deep by 24 Inch-Diameter Pool of Molten U-3/4 wt% Ti Alloy. (Reference 114)
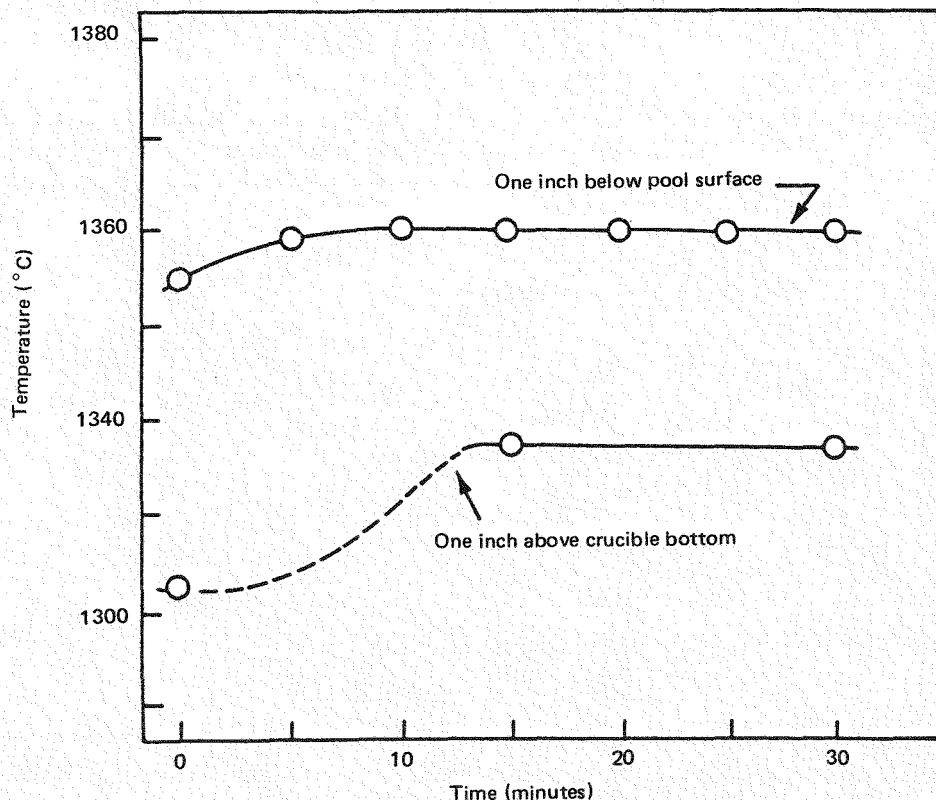
large crucible of molten U-3/4 wt% Ti alloy. From this data, it was evident that selective induction heating was occurring in the crucible. Addition of a graphite ring at the crucible base proved beneficial in reducing the effect. The size of the molten pool in this study was 10 inches deep by 24 inches in diameter. Temperature measurement was with Pt, Pt-Rh immersed thermocouples. At equilibrium, the measured temperature difference between top and bottom of the melt was about 25 °C.

The ΔT of 25 °C is not large, but it can cause segregations in hydrogen contents. At 10-micron pressure, the hydrogen solubility as a function of temperature changes significantly. A 25 °C difference could account for a difference of about one ppm hydrogen between the two temperatures. Hydrogen concentrations above the solubility limits would form hydrogen gas, which should rise from the melt.

Mold Heating

When uranium is poured into a cold mold, considerable cold shutting and surface porosity result. Increasing the pour temperature and rate of pour of uranium into a cold mold reduces cold shutting and surface porosity, but the defects remain at unacceptable levels.

To correct these casting defects, it is necessary to to heat the mold. Optimum results are obtained when the mold bottom is 700–850 °C and the mold top is 1050–1150 °C. This heating of the mold not only eliminates cold shuts and surface porosity, it also promotes directional solidification and eliminates excessive piping.

Another advantage of heating the mold to at least 700 °C is that it completely removes water from the coating. It had been established that 700 °C is the minimum temperature for complete water removal. If the water is not removed, its reaction with uranium produces gases, which form pits on the surface. A further advantage of heating the mold is that it allows for nominal (i.e., lower) superheating in the melt, and this reduced temperature inhibits breakdown of the crucible coating, thus minimizing carburization of the melt.

In a single-inductance coil furnace, the molds are positioned with their tops in the coil and the bottoms out of the coil. Trial and error determines the exact position for a new crucible-mold combination and, once found, determines and fixes the stack height for future melts with that crucible-mold combination.

Directional Solidification

Heating of the mold, coupled with the 300–400 °C temperature gradient from bottom (750 °C) to top (1100 °C) of the mold, promotes directional solidification. This is extremely important in the casting of uranium. Most of the heat flow from the mold should be through the bottom. To promote this, graphite blocks replace firebricks in the underlying refractory stack supporting the mold. Freezing should be from the bottom to the top of the mold via a solid-liquid interface as horizontal or flat as possible. This freezing pattern allows excellent drossing of undesirable solid and liquid impurities as well as gaseous impurities. Fortunately, most undersirables are lighter than uranium, and the buoyancy force to escape is quite large. The flat or planar freezing interface spreads the dross over a large area and practically eliminates the presence of a shrinkage pipe and its conical or V-shaped collection of garbage. The conical shrinkage pipe is, of course, due to a nonplanar, or conically shaped advancing solid-liquid freezing interface. The extreme of this is a vertical seam of defects at the center of the ingot caused by the freezing interface being near vertical, i.e., too much heat flowing from the sides of the mold.

Crucible/Mold Dressing

When new crucibles, bungs, stopper rods, stirrers, and molds are received from the graphite shop, they are prepared in the following manner. The surfaces of the graphite components that will contact the molten uranium are hand rubbed with a water-soaked rag until the surface luster dulls. A "watery" mixture of yttrium oxide (Y_2O_3) is hand applied in a thin coat and allowed to air dry. The surfaces are painted with a yttria wash of normal consistency using a brush, roller, or sprayer and air dried.

The yttria (Y_2O_3) wash of normal consistency is prepared in the following manner. First, 2000 grams of yttria are combined with 2000 mℓ of tap water and 60 grams of sodium CMC (sodium carboxy methyl cellulose). The solution is mixed and milled for 48 hours immediately prior to application. Mixing is in a rotating "barrel-type" ceramic ball mill.

After casting, the skull and castings are removed from the crucible and mold, respectively. The skull along with all uranium fines are transferred to an "old" crucible in a "burn-out" hood. The metal skull is ignited and burned to oxide.

The used crucible, mold, stirrer, stopper rods, and bungs are cleaned with a vacuum hose to remove the loosely adhering material. The remainder of the coating in the liquid metal zone is removed using a water spray and hand scraping or by using a fine- to medium-grit sandpaper and wiping gently by hand. This is followed by a final cleaning with the vacuum hose. Yttria, compared with other mold washes, is relatively easy to strip after a casting run. The bung hole and stopper rod are then reshaped and sized with hand tools to obtain proper seating. Finally, the crucible, mold, bung, stopper rod, and stirrer surfaces that are to contact the molten uranium are painted with a yttria wash of normal consistency using a brush, roller, or sprayer and air dried.

Ingot Dressing

After the filled molds are removed from the furnace, the ingots are broken out by stripping away the split mold. The molds are returned to the mold dressing station for reconditioning and re-use. For handling purposes and minimizing air oxidation, the ingots may be immersed in water after breakout for accelerated cooling. The ingots are then weighed and casting yields determined. Extrusion and rolling billets ingots normally weigh 150 to 700 kg each with casting yields of 95 to 98%.

Dressing the ingot may include removal of surface protrusions with a hand-held grinder or chipper in an enclosed and ventilated space. However, in most cases ingots have no objectionable surface protrusions and do not require this treatment. Nevertheless, if molds are used past their prime, protrusions at chipped seams occur. Sometimes, it is economical to use molds past their prime unless surface finish in the as-extruded or as-rolled product is of utmost importance.

The next dressing operation is sawing off the top part of the ingot. The top is the last metal to freeze and contains the shallow shrinkage pipe. The top part of the casting is also high in drossed impurities. Generally, cropping of one to two inches from the highest point is sufficient metal removal. After visual observations, additional crops may be taken to remove hot-top defects. This top-crop, sometimes referred to as a "hot-top" is often directly recycled by blending into the charge for the vacuum casting operation. If ingot purity is paramount, low-purity scrap blending may not be allowed, and the hot-tops are consolidated by vacuum remelting and used in other lower-purity metal applications.

Reciprocating power hacksaws, circular milling cutter saws, or abrasive cutoff wheels are used for top-croping uranium ingots. With hacksaws and cutoff wheels, the kerf is relatively narrow (about 1/8 inch) and the finely divided sawdust fines are pyrophoric and not directly recyclable. The kerf of the milling cutter is wider (about 3/8 inch), but produces heavier cuttings (chips or turnings), which can be directly recycled. In all cases, a water-soluble oil is used as a lubricant, coolant, and rust protectant. Also, sludge from the abrasive cutoff wheel, fines from the hacksaw, and chips and turnings from the milling cutter are stored under water. The fines and sludge can easily self-ignite if allowed to dry out. The cutoff wheel sludge is disposed of by ignition in a burn-out hood, followed by burial or chemical recycle.

The fines from the hacksaw are disposed of by incinerating in a chip roaster followed by burial or chemical recycle. The chips from the milling cutter can be cleaned, briquetted, and recast directly. Use of oil base lubricants for cutting is discouraged because of their pyrophoricity coupled with the pyrophoricity of uranium during cutting. However, metal chip recycle yields are

higher with oil-based cutting coolants such as mineral oil.

The reciprocating power hacksaws cut slowly, and blade life is short because uranium is relatively hard and work hardens significantly. However, most ingots produced at Rocky Flats are top-cropped in this manner. A circular saw would be recommended for high-volume production; it cuts faster and has much longer life because the blade can be reworked and reused many times. However, the cost of a circular saw is considerably higher than that of a reciprocating saw.

Cylindrical ingots to be extruded are top cropped to obtain a flat surface for extrusion. Not all rolling ingots are top cropped. Non-cropped ingots can be rolled directly, and the contaminated top part can be recognized by the trained eye, sheared off, and recycled in the same blending manner as hot-tops. Non-cropped ingots show more camber in the rolled product than cropped ingots; and if this is a problem, the rolling ingot is top cropped.

Chemical Sampling and Analyses

Every heat at the Rocky Flats Plant is accompanied by a minimum of one chemical analysis. A typical

chemical analysis for a multiple cast slab ingot is listed in Table 2. The chemistry of all vacuum-induction cast ingots are within specifications more than 96% of the time. Ingots that are out of specification are sawed up and blended with higher-purity material to obtain "in-spec" ingots.

Machine turnings are used for most chemical analyses. When only one analysis is made per heat, the turnings are taken from the top part of the ingot. When rolling ingots that are not scalped, the turnings are taken from a one-inch hole drilled in the top corner of the ingot at the 1/2-inch depth level. The location of the drill hole is such that it is part of the top crop or part of the shear scrap of rolled plate, sheet, or strip; thus, it does not subtract from usable metal. In extrusion ingots that are scalped after casting, the turnings are taken from the upper surface of the top-cropped cylindrical ingot. About 25 grams of coarse turnings are placed in a plastic bag, which is marked with the ingot number and sent to the in-house laboratory for analyses. In multiple ingot castings, only one ingot is normally sampled. Experience shows that sampling in the above manner is representative of all usable metal in the sister ingots from one pour.

TABLE 2. Typical Ingot Chemistry for a 99.85 wt% Uranium Stream

Test	Actual Analyses		Specifications
	Average Value	Spread	
Density	18.90 g/cm³	±0.01	>18.75 g/cm³
Uranium	99.90 wt%	±0.05	<99.85 wt%
Carbon	65 ppm		<150 ppm
Fluorine	– ppm		ppm
Hydrogen	2 ppm		<1 ppm
Oxygen	40 ppm		<100 ppm
Nitrogen	15 ppm		<40 ppm
Aluminum	25 ppm		<30 ppm
Copper	<10 ppm		<50 ppm
Iron	50 ppm		<100 ppm
Nickel	15 ppm		<50 ppm
Silicon	45 ppm		<100 ppm
Magnesium	<10 ppm		<30 ppm
Molybdenum	<10 ppm		<100 ppm
Niobium	<10 ppm		<50 ppm
Titanium	<10 ppm		<50 ppm
Zirconium	<10 ppm		<50 ppm

Hydrogen analysis is generally not done at the ingot stage because subsequent molten salt bath processing and vacuum heat treating can significantly change the hydrogen content. If hydrogen analysis is done at the ingot stage, a machined 1/8-inch cube is taken from the upper part of the ingot.

Ingot Inspection

After mold breakout, the ingots are visually inspected. For Rocky Flats ingots, the shrinkage pipe for single and multiple castings is very shallow. External measurements of the visual shrinkage pipe are nearly always less than two inches and generally less than one inch. On multiple castings, the height of individual castings is normally within one-half inch of each other, indicating very equal filling of the individual cavities. The leveling of the crucible on the furnace stack aids in the equal filling of molds. External inspection of the ingot surfaces generally reveals no cold shuts, porosity, or striations. The only external defects normally observed are minor seams mirroring the split mold junctions and some minor scabbing at the bottom of the mold where the liquid metal first impinged.

No direct effort is made to observe the interior of routine castings. However, before a casting becomes a routine one, 1/4-inch-thick vertical and horizontal sections are sawed and milled from the development as-cast ingot. The sections are polished and macro-etched to show freezing pattern, internal structure, prior gamma grain size, and internal porosity. Generally, no internal porosity is observed. The prior gamma grain size of castings is large and does not show a strong preferred orientation. Prior gamma grains are larger than ASTM 00 and sometimes as large as one to two inches in diameter. A slow cooling rate and high purity promote a large prior gamma grain size. The prior gamma grains are generally equiaxed in shape. The vertical and horizontal sections are also radiographed to verify freezing patterns and to check for internal porosity, i.e., pores greater than about 20 mils in thickness, the detection limit of this geometry. Generally, no porosity is observed radiographically or on the etched surfaces.

As a check for microscopic interdendritic porosity, small one-inch sections are given metallographic polishes and observed at high magnification in the as-polished and etched condition. Generally, no interdendritic porosity is observed. Whole flat rolling ingots are sometimes radiographed nondestructively to check for gross centerplane porosity; occasionally, whole cylindrical ingots are nondestructively radiographed to check for gross centerline porosity.

In summary, Rocky Flats Plant ingots are generally free from surface and internal defects. Visual inspection of the ingots by a trained foundryman and a trained pressman provides sufficient control to certify ingots against surface defects prior to rolling or extrusion. Ingots having harmful defects should not be processed; instead, they should be remelted and recast. Cast ingots should be held in storage and not be released for processing until their chemistry is certified by complete analysis. Nondestructive radiographing and nondestructive ultrasonic inspection to spot check for gross centerline or centerplane porosity are proven quality control tools, but neither is performed routinely at Rocky Flats.

Alloying Techniques

There are several techniques to alloy uranium by induction melting. To a large extent, the method selected depends upon the desired alloy homogeneity and impurity content.

Direct additions of the alloying ingredient require longer hold times for dissolution into the molten uranium. If the protective coating deteriorates under these conditions, a reaction of the uranium and alloying elements with the graphite occurs. After forming a carbide, elements such as titanium and zirconium float to the top of the melt because of their lower densities. This causes a loss in total alloy content and nonuniform chemistry from top to bottom within the ingot.

The greater the surface area of the alloying element, the shorter the time required for its dissolution; hence, there is less loss from carburization. For dilute uranium alloys of titanium or zirconium, sponge (available commercially) has been successfully used. During crucible charging, the sponge is placed near the bottom, allowing flotation

because of its lower density. Difficulty in achieving a uniform alloy results if the sponge is placed on or near the top of the charge where it cannot mix with the bottom of the melt.

Alloying with niobium presents a formidable problem because of its higher melting point. Using thin sheet or powder fails to prevent considerable losses during melting. For this reason, prealloyed material is used. Prealloying can be accomplished by arc melting a higher-composition ingot, which is then diluted back in the crucible with uranium by vacuum-induction melting.

Alloying can be done during the metal reduction stage. This procedure has been used successfully with molybdenum, niobium, and zirconium. Here again, a higher composition can be made and diluted during VIM refining and casting.

A summary[76] of typical vacuum-induction melting parameters at the USDOE Y-12 Plant and FMPC Plant is presented in Table 3 along with resultant alloy chemistry. At Rocky Flats, the U-3/4 wt % Ti alloy is made using titanium sponge with a 5% overage and yttria crucible-mold washes. The same parameters are used for unalloyed uranium. The U-2.2 wt % Nb alloy is made by prealloying and then diluting with uranium. A U-6 wt % Nb alloy is arc-melted and then diluted during vacuum-induction melting in yttria-coated graphite containers. Plunge stirring is used at 15-minute intervals and immediately prior to pouring. For small-mold diameters, excellent ingot homogeneity is achieved.

Published articles relating to the vacuum-induction melting and casting of uranium alloys are:

U-Al (References 77, 124, 125)

U-C (Reference 77)

U-Cr (Reference 126)

U-Mo (References 77, 79, 125, 127)

U-Nb (References 77, 79, 102, 128-131)

U-Si (References 74-75, 77, 84, 91, 100, 104-107, 111, 116-117, 125, 128-129, 132, 141)

U-Ti (References 77, 95, 101, 125, 130)

U-V (Reference 125)

U-Zr (References 77, 104, 129, 132)

URANIUM VIM CASTING INDUSTRY

Main Products

In the manufacture of wrought uranium and uranium alloy products, it is most often necessary to produce intermediate forms such as flats, rounds, and tubes. The flats are strip-rolled from flat ingots, and the rounds are extruded or rod-rolled from round ingots. Tubular products are made from back-extruding round ingots, by forward-extruding and piercing round ingots, or by forward-extruding hollow round ingots over a mandrel. In the manufacture of uranium flat, round, and tube stock, a cast ingot is the initial shape. The intermediate flat, round, or tube shape often undergoes further processing such as drawing, bending, swaging, and flaring before being machined to the end product. The subject of this report is the proper manufacture or casting of the initial rolling or extrusion ingots and, in particular, those that are vacuum-induction melted and cast. This is a major step in the overall manufacture of most finished uranium products.

Several factors normally prevent the direct extrusion or rolling of derbies to flat, round, or tube stock. Internal and external porosity, internally and externally trapped slag, and dissolved gases lead to surface and internal defects in the rolled or extruded flat, round, or tube stock. A vacuum-induction melting of the derbies will eliminate most porosity, slag, and dissolved gases. The VIM operation is a necessary metal refining step in the manufacture of high-quality uranium metal products. VIM casting operations also yield a shape more amenable to rolling and extrusion. In addition, it allows direct blending and consolidation of metal scrap.

Overview

An industry overview of depleted uranium melting and casting capabilities is depicted in Figure 13. It compares VIM to other uranium melting and alloying operations, such as vacuum-arc melting (VAM), electron-beam melting (EBM), skull melting, electroslag melting, continuous casting, and alloying by cold reduction. For clarification purposes, VIM is normally used in continuous casting, and

TABLE 3. Typical Induction Melting Practices at the USDOE Y-12 Plant and the Feed Metal Production Center[76]

Uranium Alloy Weight Percent	Charge Material	Weight Kilograms	Crucible Coating	Mold Coating	Ingot Shape	Power Kilowatts
U-3/4 Ti*	Derby Uranium, Titanium Sponge, and Recycled Uranium	360	Plasma-Sprayed ZrO_2	Zirconite Mold Wash (A)	Slab 4 × 20 × 20 in.	175
U-0.65 Zr*	Recycled Uranium Zirconium Sponge	15	MgZrO Brush Coated	MgZro$_3$ Wash	Slab 1 × 5 × 7 in.	45
U-10 Mo*	Derby Uranium and Molybdenum Powder	1055	Plasma-Sprayed Yttria-Stabilized ZrO_2	$MgZrO_3$ Wash	Slab 8 × 20 × 20 in.	175
U-6 Nb*	Prealloyed by Arc-Melting	375	Plasma-Sprayed ZrO_2	Zirconite Mold Wash (A)	5 in. Diameter 42 in. Long	175
U-8 Mo-1/2 Ti*	Derby Uranium, Titanium Sponge, Molybdenum Powder	164	Plasma-Sprayed ZrO_2 ZrO_2	$MgZrO_3$ Wash	7 in. Diameter 14 in. Long	60
U-3/4 Mo-3/4 Zr-3/4 Nb-1/2 Ti*	Derby Uranium, Titanium, and Zirconium Sponge, Molybdenum Powder, Niobium Chips	360	Plasma-Sprayed ZrO_2	Zirconite Mold Wash (A)	Slab	175
U-5 Zr**	Co-reduction UF_4+ Ca (4% excess) + Zr Fines (100% excess)	230	$MgZrO_3$ Wash	$MgZrO_3$ Wash	5 in. Diameter	200
U-5 Mo** (Double Melt)	Co-reduced UF_4 + Mg (4% excess) + Mo Powder + MoO_3 (50% excess)	230	Zr Crucible for First Melt. Second Melt BeO Wash	$MgZrO_3$ Wash	5 in. Diameter	200
U-(ppm Additions)** 1200 C, 300 Ni, 500 Fe, 150 Cr, and 250 Si	Alloying materials Charged as Powders and/or Chips	545	Not Coated	Not Coated	7 in. Diameter	200
U-0.075 Al**	Derby Uranium, Aluminum Pellets (Al in Mold)	– – –	Not Coated	Not Coated	13 in. Diameter	200

* Y-12 Plant, Oak Ridge, TN
** Feed Metal Production Center, Fernald, OH

TABLE 3. Typical Induction Melting Practices at the USDOE Y-12 Plant and the Feed Metal Production Center[76](Cont'd)

Uranium Alloy Weight Percent	Frequency Cycles/Sec	Melting Cycle	Mold Temperature (°C)	Typical Chemical	
				Top	Bottom
U-3/4 Ti*	960	1370 °C-Cool to 1320 °C-Hold 30 Minutes-Pour	1000 °C Top min. 700 °C Bottom	0.785 wt% Ti; 30 ppm C	0.756 wt% Ti; 25 ppm C
U-065Zr*	960	1310 °C-Hold 10 Minutes-Pour	– – – –	0.60 wt% Zr	0.58 wt% Zr
U-10 Mo*	960	1450 °C-Hold 30 Minutes-Pour	360 °C Top 225 °C Bottom	10.59 wt% Mo; 88 ppm C	10.31 wt% Mo; 87 ppm C
U-6 Nb*	960	1450 °C-Hold 30 Minutes-Pour	1000 °C Top min 700 °C Bottom	5.94 wt% Nb; 79 ppm C	5.96 wt% Nb; 87 ppm C
U-8 Mo-1/2 Ti*	960	1425 °C-Hold 15 Minutes. Cool to 1350 °C-Hold 15 Minutes-Pour	– – – –	7.96 wt% Mo; 0.85 wt% Ti; 31 ppm C	7.364 wt% Mo; 0.666 wt% Ti; 8 ppm C
U-3/4 Mo-3/4 Zr-3/4 Nb-1/2 Ti*	960	1425 °C-Hold 15 Minutes-Pour	– – – –	Mo 0.84 Nb 0.68 Zr 0.80 Ti 0.58	C 29 ppm
U-5 Zr**	3000	1370-1425 °C-Hold 1-1/2 Hours-Pour	– – – –	5.23 wt% Zr; 50 ppm C	4.72 wt% Zr; 50 ppm C
U-5 Mo** (Double Melt)	3000	1260 °C-Long Hold-Pour/Second Melt 1370 °C-1-1/2 Hour Hold-Pour	– – – –	Carbon less than 500 ppm	
U-(ppm Additions)** 1200 C, 300 Ni, 500 Fe, 150 Cr, and 250 Si	3000	1400 °C- 1 Hour Hold-Pour	– – – –	C-1120 ppm, Ni-330 Fe-530 ppm, Cr-160 Si-270 ppm	
U-0.075 Al**	3000	– – – –	– – – –	Reported as Excellent Ingot Homogeneity	

*Y-12 Plant, Oak Ridge, TN.
**Feed Metal Production Center, Fernald, OH.

*Large Capability L - Limited Capability S - Special Use Capability

Melting/ Casting Procedure	FMPC (Westinghouse)	RFP (Rockwell)	Y-12 (Martin-Marietta)	LANL (Univ. of CA)	LLNL (Univ. of CA)	Sandia	(Western Elec.)	NMI/CM	TNS (Aerojet)	MS	(Chamberlin)	ERL (Crown Co.)	BNLL (Crown Co.)	UK	FR	USSR	CH	IN	PK	IS	SA	AR
	USA — Government							USA — Private				CAN										
VIM	*	*	*	L				*	*			*		*	*	*	*	L	L	L	L	L
VAM		*	*	L			L								L							
EBM		L	S	S	S							(*)										
Coreduction	L											L	L									
Continuous Skull Electroslag		L / * / *																				

FIGURE 13. Industry Overview of Depleted Uranium Melting and Casting Capabilities

alloys prepared by co-reduction are refined and reshaped by VIM. Tonnage-wise, VIM accounts for well over 90% of the uranium melted and cast in the world to date with VAM accounting for most of the remainder.

Melting and Casting Costs

Table 4 indicates approximate full recovery costs for melting and casting a 1500-lb heat of uranium

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TABLE 4.   Estimated Full Recovery Costs for Vacuum-Induction Melting and Casting a 1500-Pound Heat of Uranium

	Dollars Per Pound of Uranium
Direct Labor (12 hours per heat)	0.50
Direct Materials (mostly crucible/molds)	0.30
Utilities	0.15
Maintenance Labor and Materials	0.10
Equipment Depreciation	0.05
Building Depreciation	0.05
Scrap Handling	0.35
Total	1.50

consisting of a charge of 50% virgin metal and 50% solid scrap. Melting and casting a 3000-lb heat would roughly cut the cost per pound in half.

SUMMARY

Vacuum-induction melting is a widely used technique for preparing castings of uranium and its alloys. It provides several advantages, such as simplicity of equipment and operation, considerable accumulated experience, and reasonable economy. Its chief disadvantage is potential carbon contamination from the graphite crucibles used throughout the industry.

In the United States, the uranium VIM equipment consists of either a water-cooled steel shell or a micarta shell surrounding an induction coil lined with a high-temperature refractory such as alumina. In some instances, heat shields are used to prevent excessive heatup of the outer furnace shell. It is also advantageous to have separate coils, controlled with a saturable core reactor, to provide specific zone heating of the crucible and mold. The Rocky

Flats Plant uses a water-cooled steel shell, alumina brick, and makes do with a single coil to heat the crucible and mold.

In the United States, power to the coil is supplied by a motor generator or a solid-state converter where power and frequency depend upon the size of melt to be made. Frequencies of 1000 to 3000 cycles per second are commonly used to melt uranium, with 1000 cycles being the most common.

For uranium and its alloys, 60-cycle low frequencies are sometimes used in conjunction with higher frequencies to provide some electromagnetic stirring of the melt. Most of the melt heating is conduction and radiation from the inductively heated graphite crucible, which is an excellent susceptor material. Rocky Flats uses 960 Hz supplied by solid-state converters.

Vacuum systems enable evacuation of the chamber to less than 100 microns with ample pumping capacity to maintain less than 200 microns during melting. Systems with routine vacuums less than 10 microns and pumping capacities to maintain less than 50 microns during the melt are recommended and used at Rocky Flats. The vacuum equipment should consist of positive displacement and/or blower-type pumping systems rather than oil-diffusion systems.

Uranium is a highly reactive metal and, therefore, requires selection of a compatible material for the crucible and mold to avoid excessive contamination. Allowance must also be made for the reactivity of many alloying ingredients. Coated graphite crucibles and molds are used extensively in the industry. Crucible coating technology is limited, and most coatings break down to some extent at melt temperatures, especially when temperatures exceed 1400 °C, causing carbon pickup from the exposed raw graphite. The best combination of economics and performance is a crucible wash of yttria. The coating requirement for the mold is considerably less stringent than for the crucible because of the minimal molten metal contact. A mold coating should withstand high-temperature shock and have suitable mold release characteristics.

Uncoated graphite molds are sometimes used in the United States for cold mold casting, provided that a dense grade of graphite is used. For quality castings, a yttria mold wash is recommended, not only for its protective nature, but also for ease in stripping during mold reconditioning. Yttria coatings on all parts contacting molten uranium are used at Rocky Flats.

Operating procedures vary with each composition to be melted and (to some extent) with each furnace. It is, therefore, difficult to apply a set of operating conditions that will universally produce the desired results. However, operating parameters showing the greatest influence can be generalized.

In most cases, it is advantageous to melt at the lowest temperature and shortest hold time possible for any given metal or alloy. This is due to deterioration of the coatings as temperature increases. Excessive temperatures can also cause reaction between mold and ingot with resultant sticking. Inadequate temperatures require longer hold periods, causing deterioration of the coatings. In addition, temperatures too low provide inadequate diffusional alloying and also tend to produce cold shuts on the casting surface.

A superheat of 100 to 150 °C above the liquidus of the alloy is considered a good starting point; a 200 °C superheat is used at Rocky Flats. Temperatures are usually measured by an optical pyrometer focused on the melt. Because of oxide formation at the melt surface, allowance must be made for errors in the readings.

When uranium metal or alloy is poured into a cold mold, considerable cold shutting and surface porosity results. Increasing the pour temperature and rate of pour improves the cold shutting, but surface pitting may remain. Optimum results are obtained when the mold bottom is heated to 700-750 °C. This is the established minimum temperature at which the water in the graphite and coating is completely removed. If the water is not removed, its reaction with uranium produces gases that form pits on the surface. This temperature also allows for nominal superheat in the melt to prevent cold shuts on the ingot surface. The temperature at the mold top should be 300 to

400 °C higher than at the bottom to promote directional solidification and prevent excessive piping. High-pour temperatures and hotter molds also contribute to alloy segregation and formation of carbides, which may or may not float to the top of the ingot during slow cooling.

Poor atmosphere control during melting results in heavy dross formation, rapid deterioration of the graphite crucibles, and poor melting and alloying efficiencies. Cold pumpdown of the system to less than 50-torr pressure with a leak rate not exceeding 20 torr per hour is generally sufficient. The lower the pressure and the lower the leak rate, the better the melt purity.

Preliminary outgassing of the coated crucible and mold is advisable; this treatment consists of heating to 1350 °C for one hour. If the furnace has been newly bricked, an empty run up to 1350 °C is also recommended for outgassing.

Alloy segregation, which occurs in vacuum-induction melting, can be separated into segregation caused by the melting process and that resulting from inherent solidification processes. Segregation resulting from the melting process is chiefly caused by gravity segregation in the melt and more directly to carbide formation of the alloying ingredient and subsequent drossing out of the carbides. If large ingots are cast requiring relatively long solidification times, carbide flotation can continue in the mold. Alloy separation in the crucible can also occur as a result of large density differences between the alloying addition and uranium and because of thermal gradients in the melt. Sixty-cycle, low-frequency electromagnetic stirring is questionable for reducing these effects. Mechanical mixing of the melt using a paddle or inserting an argon lance into the melt is recommended for stubborn mixing problems.

The importance of directional solidification in obtaining a sound casting can not be overemphasized. To produce a sound ingot, it is critical that freezing proceed from bottom to top and not from wall to center.

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